Volume 6 MARCH, 1935 Number 3

PHYSICS

A Journal of General and Applied Physics

A Multiple Space Charge Effect

EVERETT W. THATCHER, *Union College, Schenectady, N. Y.*

(Received December 7, 1934)

An investigation has been made of certain phenomena associated with space charge external to the grid which surrounds the cathode in a multi-grid tube. This paper reports a new effect, the essential feature of which is a negative plate current-emission characteristic. The conditions under which this phenomenon is observed are outlined. Results of experiments with double and triple grid tubes are summarized. In general, two points on the characteristic may be found for which the plate current is independent of small variations in emission, whether of a statistical or "functional" nature. An explanation is presented based on the interaction of multiple space charge. An application to the problem of high gain amplification is suggested.

I. INTRODUCTORY

THE study of the effect of space charge on thermionic currents has been restricted mainly to those cases in which maximum density of electrons occurs in close proximity to the emitter. Under certain conditions, control of the current in multi-grid tubes may be produced by what we shall designate as" external" or "multiple" space charge. These terms are applied here to space charge resulting from accumulation of electrons in regions of the tube external to the grid which surrounds the cathode. One case in which this situation was used has been treated briefly by the writer in connection with studies of the shot effect.[1, 2]

The characteristics associated with external and multiple space charge have been investigated. It is the purpose of this paper to report a new effect which appears under these conditions.

II. GENERAL DISCUSSION

Space charge limitation in the conventional sense involves a saturation level beyond which the current ceases to rise with emission. The essential feature of the effect reported here is a situation in which the plate current *decreases* with increasing emission, preceded and followed by regions in which the current rises with emission. The curve which represents the plate current as a function of emission will have two horizontal tangents and a region of negative slope.

Experiments with double and triple grid tubes have shown that the conditions favorable to the appearance of this effect include:

(1) An accelerating field for electrons as they leave the emitting surface. This is maintained by holding the first grid at sufficiently high positive potential to *prevent* space charge limitation of the current from the cathode. This also serves to prevent the escape of positive ions, which are usually, if not always, present in thermionic emission.[3]

(2) A retarding field in which the stream velocity is reduced before passing into the outer space. This is produced by making the second grid only slightly positive with respect to the cathode. It may then be considered as a virtual emitter for the outer space.

[1] E. W. Thatcher and N. H. Williams, Phys. Rev. **39**, 474–496 (1932).

[2] E. W. Thatcher, Phys. Rev. **40**, 114 (1932).

[3] Reference 2, p. 479.

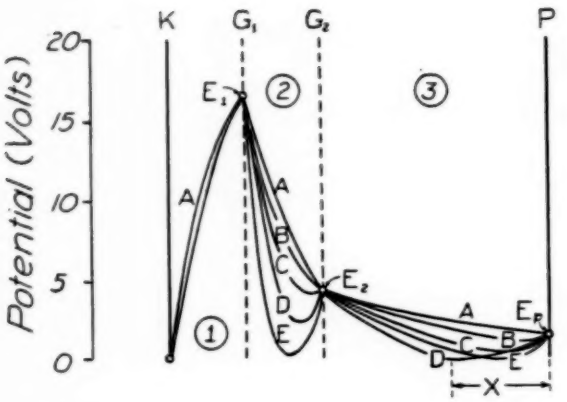


FIG. 1. Representation of element and space potentials in double grid tube (longitudinal half-section). *A*, *B*, *C*, *D* and *E*, indicate successive stages in the establishment of multiple space charge.

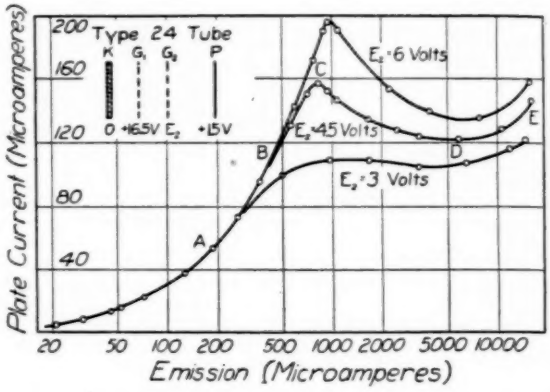


FIG. 2. Multiple space charge characteristic in double grid tube (Type 24). Plate current is plotted against emission (logarithmic scale) with constant grid and plate potentials. The three curves represent three outer grid potentials.

The situation may be discussed most easily by considering the results of experiments with a double grid tube (Type 24). The emitter in this tube is of the indirectly heated type which furnishes an equipotential cathode, thus avoiding the difficulty presented by the drop in potential along a filament. The 24 also has relatively large spacing between the second grid and plate, a situation which is favorable for the establishment of space charge in this region of the tube.

III. Multiple Space Charge in the Double Grid Tube (Type 24)—Results and Interpretation

Fig. 1 shows a longitudinal half-section view of the elements in the 24 tube. K refers to the cathode, G_1 to the first grid, G_2 to the second and P to the plate. A set of element potentials under which external space charge effects are observed is indicated by the position of the points E_1, E_2 and E_p taking the cathode potential as zero. If we neglect penetration and end effects, we may represent qualitatively the form of the inter-electrode fields by drawing lines connecting these points. The cylindrical arrangement of the elements leads to a space potential distribution of the type shown by lines A.[4] The field at any point can be obtained from the slope of the potential curve.

When emission occurs, all space potentials are depressed, the largest effect occurring in the region of weakest field (i.e., near the plate). With increase in emission, a cylindrical surface of minimum potential is established near the plate which travels inward toward G_2 as the electron density grows. It is assumed that successive stages in this process might be indicated by such curves as B, C and D.

Fig. 2 shows the variation in plate current with emission for several values of E_2. For convenience one of these curves ($E_2=4.5$ volts) has been divided into sections. The letters which mark the divisions are placed to represent the situations indicated by the same letters in Fig. 1. It is characteristic of the curves shown that the plate current first rises to a rather sharp maximum level determined by the grid potentials; then, with no change in condition except a further *increase* in emission, falls, passes through a broad minimum and eventually rises slowly. While interest attaches primarily to the current which reaches the plate, our understanding of the situations which affect the electron stream is materially aided by a knowledge of the grid currents. Fig. 3 gives the relative magnitudes of these currents expressed as fractions of the total emission. The case when $E_2=4.5$ volts is taken as an example.

The currents to all three electrodes are limited originally by the number of electrons present (i.e., by emission). Let us now follow the changes which occur as the emission increases. At B, Fig. 3, we note an increase in the fraction of the stream reaching G_2, and a corresponding de-

[4] See Chaffee, *Theory of Thermionic Vacuum Tubes*, p. 75.

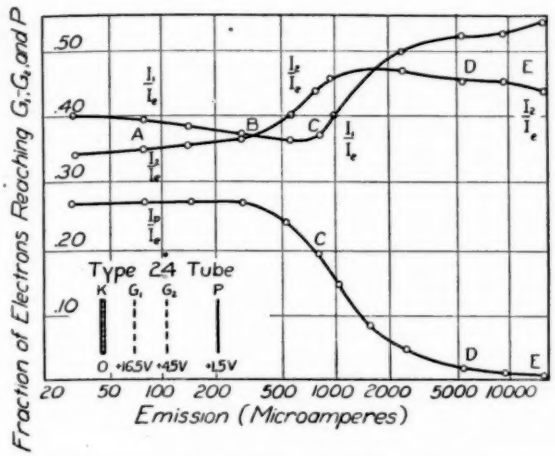


FIG. 3. Relative number of electrons reaching the elements as emission changes.

crease in I_p/I_e. This is interpreted as an indication that space charge conditions have been established in the outer space. If this is correct, limitation from this cause will retard the increase of current to the plate. This stage is evident in Fig. 2 in the decrease in slope of the plate current curve.

A simple space charge effect might be expected to produce a saturation current level, the height of which would depend largely on plate potential. In place of this we find the current reaching a maximum and then declining with increased emission. Can this phenomenon be explained on the basis of space charge?

Let us consider the influence of the surface of minimum potential which appears to be established in space 3. Only those electrons having sufficient velocity to move against the retarding field as far as this surface will contribute to the plate current. Those of lower speed will return in the direction of G_2. They may strike this grid, or pass through its mesh toward G_1. Their behavior will depend on the fields encountered in space 2. In any case their presence near G_2 will further lower the space potential in the vicinity. (Unless all returning electrons strike the grid on the first passage, this influence may extend to the potential of space 2. The effect is intensified if several passages through the mesh occur before capture by G_2.)

The maximum current to G_2 is not reached until the emission is about 2000 microamperes. At 800 microamperes the curve showing I_1/I_e makes a sharp bend upward, because, it would seem, of the return of space charge limited electrons from a minimum potential region in space 2. Since increased emission, at least in the earlier part of this stage, will mean greater electron density in space 3, the radius of the outer surface of minimum potential will become smaller. The result is seen to be an increase first of I_2 and then of I_1 at the expense of the plate current.

There remains to be explained the minimum level of I_p followed by its rise as emission becomes quite large. The potential distribution in the tube when the plate current reaches the minimum stage may have the general character of the lines marked D in Fig. 1. Neglecting, for the moment the possible contribution of change in initial emission velocities and non-radial components, we may apply the three-halves power space charge approximation to the current from the minimum potential surface in space 3. Thus, $I_p = kV^{\frac{3}{2}}/x^2$, where V is the potential of the plate with reference to the minimum, and x the distance from the plate at which the minimum is established by space charge.

Let us now consider the influence on this current of the potential minimum produced in space *2* by the increase in density of the emitted stream. Electrons which now pass through G_2 will enter space 3 with higher average velocities. It should be recalled that the current density at any point is given by $i = \rho v$ where ρ is the electron density. If the number entering space 3 remains constant (due to limitation in space 2) this can result only in a *decrease* in space charge near G_2. Hence the potential minimum region should move outward toward the plate, and with decreasing x an increase in plate current should follow. The writer's interpretation of this change is indicated at E in Fig. 1. The actual effect on the plate current is shown at E in Fig. 2.

The influence of the potential of the second grid is clearly shown in Fig. 2. The curves for various potentials branch off from the early part of the curve which is, within experimental error, common. It seems significant that with increasing E_2 the peak shows only a slight shift toward higher total emission, while the amplitude

of the effect, i.e., the spread between maximum and minimum values of current, grows larger. Both of these trends are in line with the explanation given above. For example, a higher potential difference between G_2 and P will require a greater stream density to produce space charge limitation. Once established, however, a larger fraction of the total will finally reach G_2. This has been verified by the comparison of the curves in Fig. 3 with similar curves obtained with different values of E_2.

IV. Effect of Cathode Space Charge

It has been assumed in the foregoing that space charge limitation in the space between the emitter and first grid was not present when the phenomenon was observed. This has been shown experimentally to be true in the range thus far considered. It is reasonable to inquire into the influence of E_1 on the observed behavior of the electron stream, and in particular to test the effect of inner space charge.

When E_1 is varied, two significant effects are observed (see Fig. 4): (1) For lower E_1 the peak becomes larger and suffers a displacement in the direction of the lower total emission; (2) the final rise of plate current occurs earlier (at lower emission values) for low than for high E_1. Any change which results in more readily establishing space charge in space 2 will cause the second of the trends. This may also be said for space charge surrounding the emitter. This probably accounts for the large difference observed between the 6 volt and the 16 volt curves.

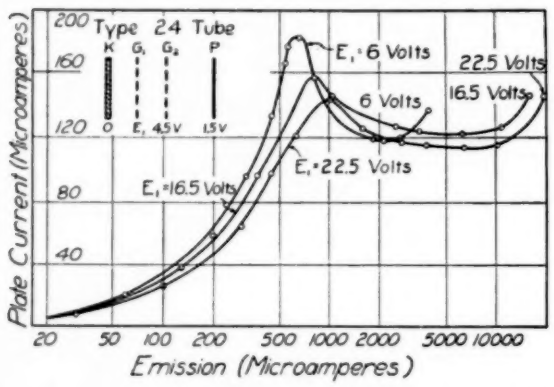


Fig. 4. Multiple space charge characteristic in double grid tube. Effect of E_1.

The displacement of the peak presents a more difficult problem. The explanation which seems probable at the present is that non-radial components of velocity become more prominent for high grid potentials. An increase in the magnitude of such components would increase space charge without contributing to the plate current.

V. Discussion

Two aspects of the multiple space charge characteristics have particularly interested the writer and it seems pertinent to call attention to them here. It has been shown theoretically and supported experimentally that the fluctuation level ($\overline{E^2}$) of the normal shot effect is reduced under space charge by a factor $(\partial I_p/\partial I_e)^2$ where I_e is the emitted current and I_p that which reaches the collector.[5, 6] *For two points on the characteristic reported here this factor is zero.* The same consideration may be extended to cover small changes in emission due to variations in the heating current or changes in the emitting surface.[7] As long as these are of the relatively small amplitude usually encountered the effect in the plate circuit is very small indeed.

It has further been noted that the electron stream reaching the outer space is pure (i.e., free from positive ions). One is therefore able to establish space charge conditions in this region *without introducing current fluctuations of the abnormal type* described by Kozanowski, Williams and the writer.[8, 9]

The fluctuation level in the output circuit of a tube operated in this manner may thus be reduced practically to that resulting from thermal agitation. If, therefore, a control grid be inserted between the virtual emitter and plate, the tube may be operated as an effective triode with the unique property that the plate current is independent both of statistical and "functional" variations in emission.

---

[5] F. B. Llewellyn, Proc. I. R. E. **18**, 243 (1930).
[6] See reference 2, p. 115.
[7] E.g., the "flicker" effect which becomes prominent at low frequencies. Johnson, Phys. Rev. **26**, 71 (1925); Schottky, Phys. Rev. **28**, 74 (1926). Pearson, Physics **5**, 233 (1934) has demonstrated the effectiveness of space charge in eliminating these fluctuations in the space current.
[8] Kozanowski and Williams, Phys. Rev. **36**, 1314 (1930).
[9] Thatcher and Williams, Phys. Rev. **39**, 479–484 (1932).

It is well known that one of the limits on the useful gain of vacuum tube amplifiers is the magnitude of these fluctuations in early stages—particularly the first. The operation of the first tube in the manner described should make possible an increased signal to noise ratio in the input to the second tube.

The properties of multiple space charge seem of particular interest in d.c. amplifier applications of the electrometer type. In this connection several schemes have been devised to minimize the effect of filament variation.[10] As far as the writer is aware, however, all these have involved the matching of tubes or setting up bridge circuits by which variations in one branch are compensated by those in another. The use of the multiple space charge characteristic should reduce the complexity of these operations and increase sensitivity by elimination of one of the main causes of instability.

VI. Results with Triple Grid Tube (Type 78)

A preliminary test of this method of operation has been made in a study of the phenomenon in a triple grid tube (Type 78). The tube chosen is one with external connections to each grid. Field conditions similar to those in the preceding case were established surrounding the emitter. The third grid was used as a control element. Fig. 5 shows the plate current emission characteristics which were obtained with $E_1 = +9$ volts, $E_2 = +4.5$ volts and $E_p = +9$ volts.

Attention is directed to the displacement of the initial peak as E_3 is varied. This peak is reached earlier as E_3 is made more negative, with consequent increase in outer space charge and reduction of plate current. The relatively

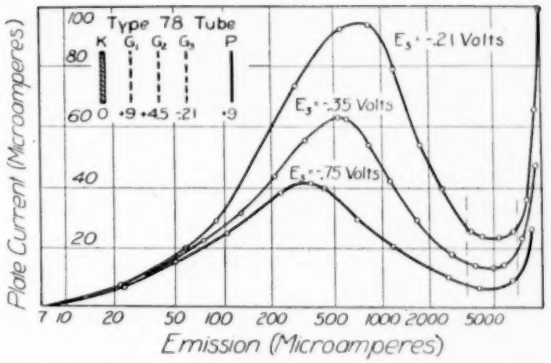


Fig. 5. Multiple space charge in a triple grid tube. Plate current *vs.* emission (logarithmic scale). Effect of control grid potential (E_3).

broad minimum occurs at about the same position on each curve. With 4500 microamperes emission, the equilibrium potential of G_3 was determined. This is the potential which the grid will assume when left free from connection with other parts of the system. If a bias of this amount is applied, no change in current will be observed.

For the tube considered here this value was found to be -0.21 volt, at which potential a stream of 23.8 microamperes reaches the plate. The transconductance in this region was found to be about 200 micromhos. With tubes designed to provide more effective control on G_3 this figure should be materially exceeded. Improvement can particularly be expected in the increase in input resistance which will result from bringing the lead from this grid to the cap, rather than through the pinch seal to the base of the tube.

VII. Acknowledgment

The writer is happy to acknowledge his indebtedness to Dr. Irving Langmuir and to Dr. A. W. Hull for valuable counsel in the interpretation of these results and for helpful suggestions in the preparation of this paper.

[10] See, for example: Soller, Rev. Sci. Inst. **3**, 416 (1932); Du Bridge and Brown, Rev. Sci. Inst. **4**, 532 (1933); Harnwell and Van Voorhis, Rev. Sci. Inst. **5**, 244 (1934); and others.

Some Electrical Properties of Ceresin Wax

A. WIKSTROM, *Cornell University, Ithaca, N. Y.*

(Received October 26, 1934)

A lower limit for the resistivity of commercial ceresin is set at 1.76×10^{19} ohm-cm if the voltage gradient is 2000 volts per cm the thickness of the sample is 0.214 cm and the temperature is 23°C. There is a comparatively large scattering of time functions used in the equation for the discharge current if expressed in accordance with Wagner's theory. The charge Q "stored" in ceresin is a function of time. At any time after the application of the voltage, this charge is proportional to the voltage. P/E was measured as a function of t. The increase is found to be more rapid than is claimed by other investigators. No variation of P/E with E was found. The results, however, are not quite conclusive, because of the inherent weakness of the method used in determining the polarization. The resistance R as defined by Richardson is calculated and found not to be constant for ceresin but to vary both with time and voltage.

A STUDY has here been undertaken of ceresin wax when subjected to a constant voltage, in order to determine some of its electrical properties.

The sample used in this investigation was obtained from a block of commercial ceresin, no attempt being made to purify the product. The reason was that this wax, which has a yellowish color, is used today to a very great extent in research laboratories as an insulator. Very little is, however, known with respect to its behavior when subjected to a constant voltage. Troubles have often been encountered due to its soakage current which prevents the conduction current from getting reasonably constant over a period of several hours after the voltage has been applied.

Apparatus and Experimental Procedure

For this investigation a test-condenser was made in the following manner. A circular brass plate (diameter = 3 inches, thickness = $\frac{1}{2}$ inch) was set into a lathe and the surface was carefully turned. The chuck was then unscrewed and laid flat on a table with the brass plate up. A paper form was made to fit the circumference of the plate and after the plate had been heated with the flame from a Bunsen burner to approximately 80°C, melted ceresin was poured onto the brass plate. The paper form prevented the wax from flowing off the plate. After 24 hours the chuck was screwed back on the lathe and the ceresin was turned down to a thickness of 0.0842 inch (2.14 mm). The surface of the wax was then covered with several layers of gold leaf such as bookbinders use. While the condenser was still in the lathe a circular cut was made through the gold to form a guard ring.

A simplified wiring diagram is shown in Fig. 1. E is a Compton electrometer of low capacity, adjusted by means of negative control to a sensitivity of approximately 10,000 mm per volt, which could be checked with the potentiometer P_1. The test condenser C_t, shown with grounded guard ring, has one plate connected with the electrometer and the other plate connected with an electromagnetic switch S_1, which made it possible to connect C_t either with the ground or with one terminal of the high tension battery B, the other terminal of which is grounded. C_s is a cylindrical condenser (see Fig. 2), the capacity of which is calculated from its dimensions using the formula: $C = L/2 \ln (d_1/d_2)$. The quantities L, d_1 and d_2 were chosen so as to make $C = 25$ cm.

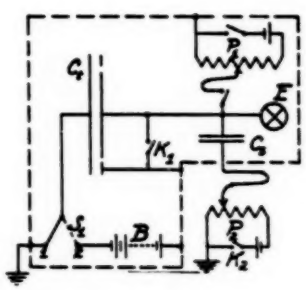


FIG. 1. Diagram of connections for the determination of the total current i_t or discharge current i_d.

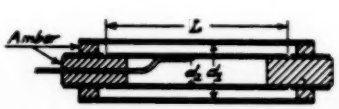


FIG. 2. Cylindrical condenser with guard rings.

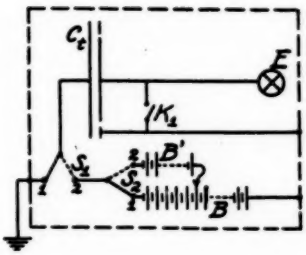


FIG. 3. Diagram of connections for the determination of the polarization P.

The dotted lines in Figs. 1 and 3 show to what extent the apparatus was shielded. Besides this shielding the switches S_1 and S_2 were in a separate grounded metal box, as these switches were operated electromagnetically. The shielding box containing the specimen had for drying purpose a large surface dish containing calcium chloride.

The time recording was done by means of a chronograph manufactured by Wm. Gaertner and Company, Chicago. Before being put into service this instrument was adjusted and checked by means of a synchronous clock motor. The maximum error was ± 0.005 of a second per minute.

In this investigation the following quantities were measured:

(1) The total current i_t *vs.* time t for different values of voltage E of the battery B (see Fig. 1).

(2) The discharge current i_d *vs.* time t for different values of the time T during which the voltage E was applied to the dielectric.

(3) The polarization P *vs.* charging time T. One such curve obtained for each voltage E.

(1) The experimental determination of i_t

Before any measurements of the total current i_t were made, the condenser C_t was left short-circuited until completely discharged. This was done by throwing the switch S_1 into position No. 1 and closing K_1 for several days. The condenser was considered completely discharged, when the insulated electrometer quadrants upon opening K_1 would not take on a charge at a greater rate than 5×10^{-16} coulomb per second. When this was the case the total current i_t was determined as a function of time in the following manner.

At a certain instant of time $t=0$ the switch S_1 was moved into the position No. 2 (see Fig. 1), while K_1 and K_2 were closed. The current, which will then flow, consists of a transient current superimposed upon the total current i_t. Because of the magnitude of the constants of this circuit the transient current subsided within a fraction of the first second. No attempt was made to measure it.

Whenever i_t was to be determined K_1 was first opened. The electrometer then drifted and, when passing a certain division N upon the scale, the time $t=t_1$ was recorded. The electrometer was now brought back to zero and kept there by adjusting the potentiometer P_2. Towards the end of this determination the continuous adjustment of P_2 was stopped, resulting in a drift of the electrometer. When again the Nth division on the scale was passed, the time $t=t_2$ was recorded. The switch K_1 was now closed and as soon as the electrometer was back to zero, another determination of i_t could be made.

The voltage change ΔE of the potentiometer P_2 was measured with a Leeds and Northrup potentiometer. If the capacity of the standard condenser was C_s, the average value of i_t between $t=t_1$ and $t=t_2$ can be calculated from the equation

$$i_t=\Delta Q/\Delta t=C_s\Delta E/(t_2-t_1).$$

This average value was associated with the instant of time $t=(t_1+t_2)/2$. It is fully realized that this is not strictly correct. The function "current *vs.* time" may even for small values of Δt, deviate appreciably from a straight line. A method of measuring instantaneous values of rapidly decreasing currents is described in a separate paper by the author.[1] For values of $t<1$ minute this method was found desirable and generally used.

(2) The experimental determination of i_d

After a certain length of time $t=T$, the test condenser C_t was completely discharged by closing K_1 and throwing the switch S_1 into position No. 1 for a sufficient length of time. Immediately after such a condenser is short-circuited a current will flow which consists of a transient current superimposed upon the discharge current i_d. Again, no attempt was made to measure the transient. The discharge current

[1] A. Wikstrom, Rev. Sci. Inst. **4**, 612 (1933).

i_d was measured in the same way as i_t and readings were continually taken until the values were of the order of 5×10^{-16} ampere or less.

It was mentioned above that the condenser was considered to be completely discharged when, upon opening K_1, the insulated pair of quadrants did not charge up at a greater rate than 5×10^{-16} coulomb per second. Even when the condenser was left short-circuited for more than nine days this rate of charge varied in an irregular manner from day to day between 1×10^{-16} and 5×10^{-16} coulomb per second. Reversing the needle potential of the electrometer had no effect upon either the magnitude or the sign of the rate of charge accumulated which was always found to be of positive sign. Thus it was not due to a leak of charge between the needle and the insulated pair of quadrants. When the electrometer was disconnected from the rest of the circuit, it took on charge at a rate of less than 10^{-17} coulomb per second. This indicates that the charge was not released from the electrometer insulators. It was only when C_t was connected with the electrometer that the rate increased to a value within the above mentioned limits.

With the same polarity of the needle the electrometer, when insulated, always drifted in the same direction. The readings obtained were therefore either too large or too small, depending upon the direction of the current measured. The values of i_t were always so large as not to be affected much by this unidirectional error. To eliminate it as far as possible it was, therefore, necessary to repeat every i_d curve under identical conditions, with the exception of E being reversed. The correct value of i_d was then taken to be the arithmetic mean of corresponding values of the two current curves obtained in these two sets of readings.

(3) The experimental determination of *P*

Richardson's[2] definition of polarization was used and except for the mechanical manipulation the method of measuring it was identical with his method. A simplified diagram is shown in Fig. 3. Before any reading was taken the condenser C_t was completely discharged as in the case of the current measurements. At an arbitrary time $t=0$ S_1 was thrown into position No. 2, K_1 of course being closed. If now K_1 was opened, the electrometer E began to drift. The direction of this drift was noticed, whereupon K_1 was again closed. We shall call this direction of drift the positive direction. At a certain instant of time $t=T$, at which the polarization was to be determined, S_2 was changed into position No. 2, thus applying to C_t a voltage $E'<E$. Immediately thereafter K_1 was opened and the direction of the drift of the electrometer was again observed. If the polarization voltage is denoted by P, then a positive drift indicates that $E'>P$, whereas a negative drift shows that $E'<P$. This procedure was repeated, beginning every time with a completely discharged condenser, until $E'=P$ was obtained, as was indicated by no drift of the electrometer for an appreciable time after that K_1 was opened.

The switches S_1 and S_2 were operated by electromagnets. A double pole knife switch closed the circuit containing the electromagnet which operated S_1 and at the same time recorded through a separate circuit the time by means of the chronograph.

The grounding key K_1 was operated by means of a lever and a string. The string, which when pulled opened K_1, was fastened to a single pole knife switch of special design. When operated, this switch first, through an electric circuit, threw S_2 into position No. 2, and then K_1 was immediately opened by means of the string. It was mechanically impossible for K_1 to open until S_2 made contact in position No. 2.

The battery B' consisted of 1.5 volt dry cells, thus making a fine adjustment of E' possible. A potentiometer, not shown in Fig. 3, was also included for the same purpose.

In order to minimize disturbances due to temperature variations, the research room was kept at 23°C with a maximum variation of ±0.5°C.

Experimental Results

A preliminary test showed that the volume resistivity of paraffin is much higher than that of ceresin. As the sensitivity of the electrometer used in this work was not high enough to make

[2] S. W. Richardson, Proc. Roy. Soc. **A92**, 101 (1915–16).

any comparative measurements, the relative magnitude of the resistivities of ceresin and paraffin cannot be given. Strictly speaking, there is no *one* definition of "resistivity" which can be used for dielectrics in general. Various attempts have been made to define some such constant that will make it possible to apply Ohm's law to dielectrics. To the author's knowledge, so far no definition has proved successful. A typical example is dealt with later in this paper.

Data directly measured

The results of direct measurements are shown in Figs. 4, 5 and 6. These curves are taken as representative of a comparatively large amount of data accumulated during the course of the investigation. Plots of i_t against the charging time are shown in Fig. 4 on log-log paper where each curve corresponds to a different constant potential applied to the condenser. These curves are fairly straight lines from $t=0.1$ minute to $t=1$ minute.

In general the total current was only measured for 25 minutes and did not become reasonably constant during this time. In one case the current measurements were continued for 1360 minutes.

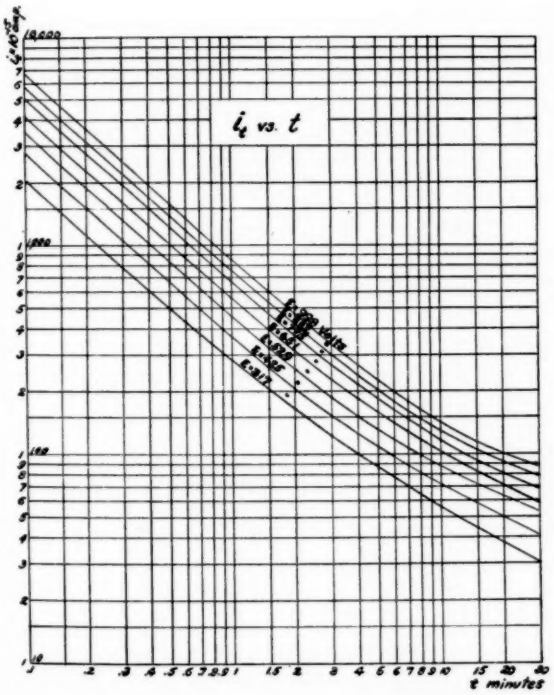


FIG. 4. The total current i_t *vs.* time.

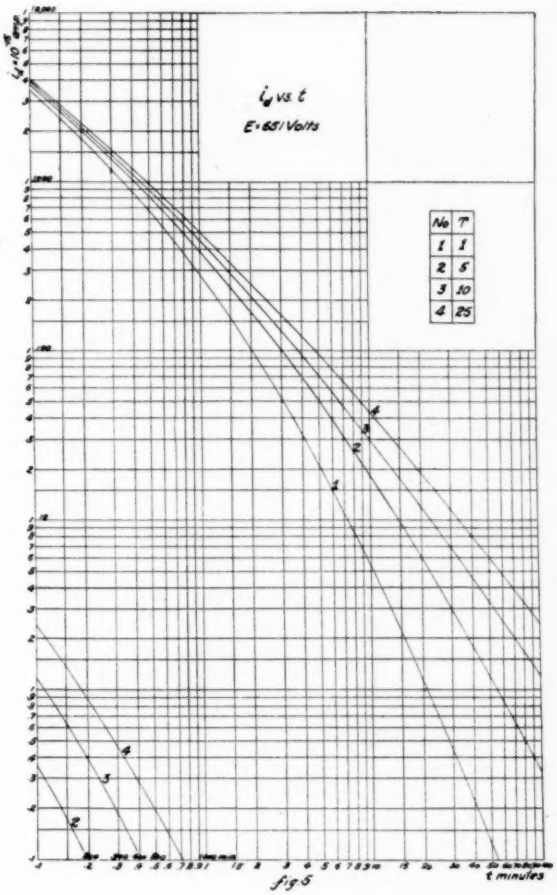


FIG. 5. The discharge current i_d *vs.* time for $E=651$ volts for four different charging times T.

The following calculations show how the resistivity ρ was determined for this particular voltage:

$$\rho = E \times \pi D^2 / i_t \times 4d,$$
$$E = 425 \text{ volts},$$
$$i_t = 4.91 \times 10^{-15} \text{ amp.},$$
$$D = 7.436 \text{ cm},$$
$$d = 0.214 \text{ cm}.$$

Hence $\rho = 1.76 \times 10^{19}$ ohm-cm.

This calculation is made in order to set a lower limit to the resistivity of ceresin wax under the following conditions: A voltage gradient of nearly 2000 volts per cm; a thickness of 0.214 cm and a temperature of 23°C. The value for ρ found by Guében[3] is 3×10^{18} ohm-cm.

The discharge currents i_d were determined for four different values of charging time T, namely:

[3] G. Guében, Zeits. f. Physik **74**, 714 (1932).

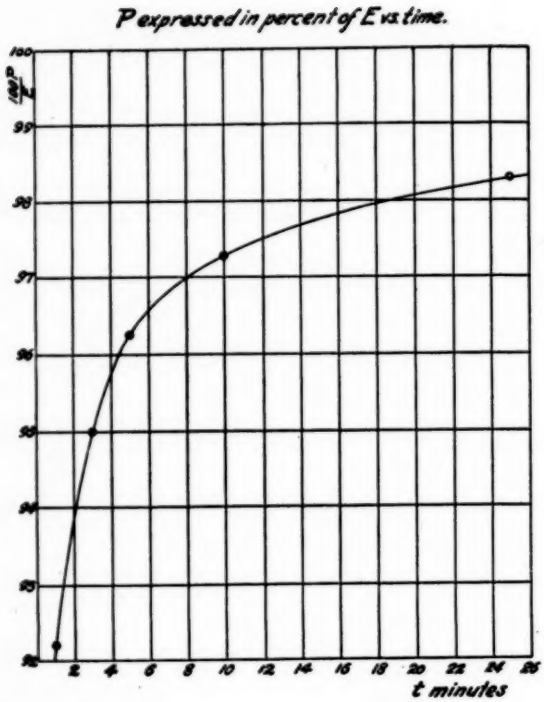


FIG. 6. The polarization P (expressed in percent of E) *vs.* time.

1, 5, 10 and 25 minutes. The curves of discharge current *vs.* time for $E=651$ volts are shown in Fig. 5 plotted on log-log paper and are seen to be slightly bent lines. As i_d decreases faster than a straight line it is possible that for very high values of t, the function $i(t)_d$ will gradually become an exponential function. It was not possible to determine whether or not this is true, as it occurs, if at all, at such small values of i_d that the accuracy of the measurements would not suffice.

The polarization P expressed in percent of E is given as a function of charging time in Fig. 6. From the author's observations there is no evidence of a variation of P/E with the applied voltage.

Data calculated from measurements

From curves shown in Fig. 5 and many similar curves obtained at other voltages, the residual charge Q in the dielectric was determined. The discharge current for a given time T of charging, is a function of the time during which the discharge takes place and if this current is expressed by the symbol $i(t)_d$, the charge Q_T, which is

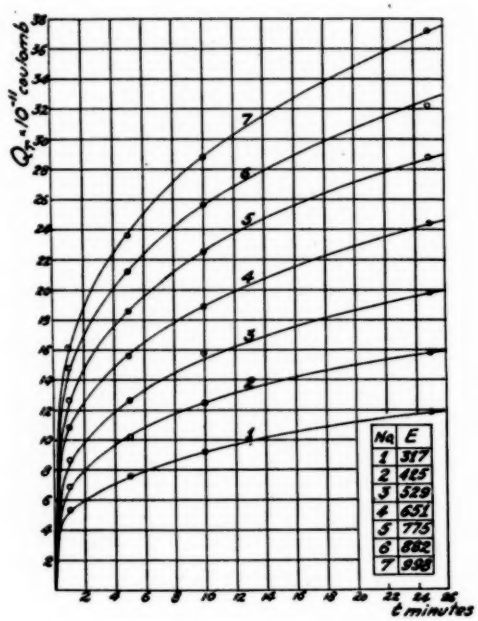


FIG. 7. Q_T *vs.* t. The charge Q_T "stored" in the dielectric *vs.* time. Each curve is for a constant potential E applied to the test condenser.

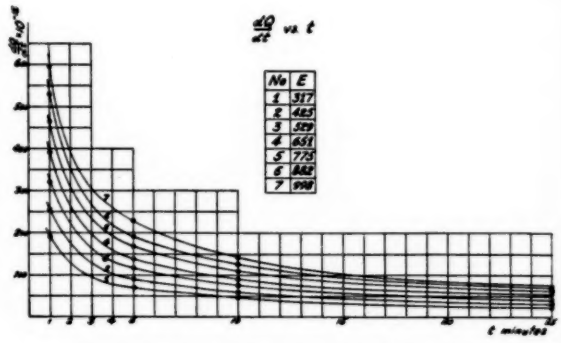


FIG. 8. The charging current dQ/dt *vs.* time.

accumulated in the dielectric during the charging time T, can be written in the following manner:

$$Q_T = \int_0^\infty i(t)_d dt.$$

As it is impractical to evaluate Q_T in a theoretical way, the discharge current $i(t)_d$ was plotted to large scale on graph paper, and the area under the curve was determined with a planimeter. The values of Q_T determined in this manner are plotted in Fig. 7 against T as abscissa. For each curve the voltage is constant.

Plotting Q_T against E for constant values of T gives the linear relations shown in Fig. 9.

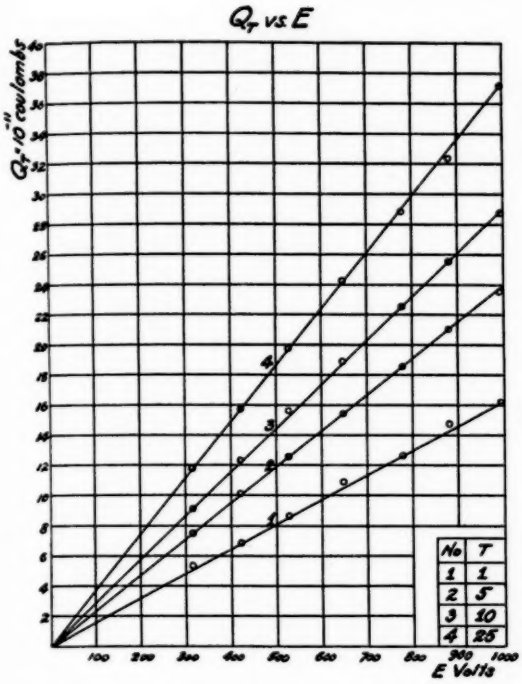


FIG. 9. The charge Q_T "stored" in the dielectric *vs.* the applied potential E. Each curve is for a constant charging time T.

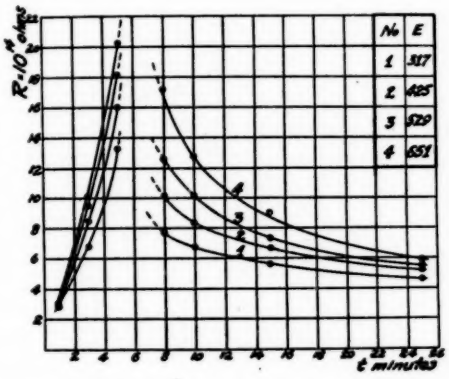


FIG. 10. R *vs.* t. The resistance R calculated from Richardson's equation as a function of time. Each curve is for a constant potential E.

The derivative of Q_T with respect to the charging time may be taken to be the charging current often spoken of as the soakage current. From the curves shown in Fig. 7 dQ/dt is plotted against charging time as abscissa. Thus a family of curves is obtained as is presented in Fig. 8. For each such curve the voltage is constant.

Since the author used Richardson's definition of polarization it was of some interest to compute the resistance R as defined by Richardson[4] in his equation for dielectrics

$$i_t = dQ/dt + (E-P)/R.$$

Fig. 10 shows this resistance *vs.* time for different voltages.

Discussion of results

The discharge curves shown in Fig. 5 are slightly bent lines. This, if compared with Wagner's[5] theory, calls for a small value of b in the equation

$$I(t) = (\epsilon k_0 b E_0 / 4\pi\theta_0\sqrt{\pi}) \int_{-\infty}^{+\infty} e^{-b^2 z^2 - z - te^{-z/\theta_0}} dz,$$

which means that there is a comparatively large scattering of the time functions, or that the time constants do not group themselves very closely round the most probable value ln θ_0.

In Fig. 6 is shown the variation of P/E with time. Owing to the rapid increase in the polarization with time and the inherent inaccuracy of the method used in determining P, we should not give too much weight to the statement that P/E is independent of E. The results shown in Fig. 6 are averages of many readings. The previous condition is, in these measurements, of the greatest importance and care was therefore exercised so as to leave the dielectric short-circuited for a long time before a new reading was taken.

While Courtener and Ignatowitsch[6] found that for ebonite P/E is independent of E, Tutschkewitsch[7] concludes that this is not true for paraffin.

A direct comparison of data for ceresin has been made possible by the work of Nasledow and Scharawsky.[8] Comparing the polarization found by these investigators with the author's values at corresponding values of time it is found that the author's values are higher. This difference may be due to different composition, temperature and thickness of the samples. Thus the difference in temperature of almost 6°C

[4] S. W. Richardson, Proc. Roy. Soc. **A92**, 41 (1915–16).
[5] K. W. Wagner, Ann. d. Physik (4) **40**, 817 (1913).
[6] A. Courtener and J. Ignatowitsch, Ann. d. Physik [5] **10**, 681 (1931).
[7] W. M. Tutschkewitsch, Ann. d. Physik [5] **6**, 622 (1930).
[8] D. Nasledow and P. Scharawsky, Ann. d. Physik [5] **5**, 429 (1930).

would, on the basis of the dipole theory as modified by Whitehead,[9] account for the quicker polarization obtained by the author. An increase in temperature decreases the coefficient of viscosity and the dipoles therefore line up faster.

The curves shown in Fig. 7 are similar to values obtained by Courtener and Ignatowitsch[6] and Nasledow and Scharawsky[8] all indicating that the charge accumulated in a dielectric approaches a maximum value of charge that can be stored in the dielectric itself. For higher values of voltage gradient it takes the same time to reach a value which is within a certain percent of Q_{max}.

The investigators referred to above have all studied samples of different thickness. It is conceivable that the thickness may have various effects upon the characteristics of a dielectric. Thus the rapidity by which the polarization takes place may possibly change contrary to the theory of similarity suggested by B. B. Kurtschatow and J. Kurtschatow.[10]

[9] S. Whitehead, Phil. Mag. [7] **9**, 865 (1930).
[10] B. B. Kurtschatow and J. Kurtschatow, J. d. russ. physik. chem. Gesell. **61**, 321 (1929).

The purpose of this investigation was to study ceresin such as it is believed to be found in use for insulation in research laboratories of today. The characteristics can probably be changed to a very great extent, depending upon the way the wax is treated while the condenser is made. In order to permit results to be compared successfully, a standard treatment should be adhered to, such treatment including the heating of the wax to the melting point for a definite time in an evacuated chamber and possibly in a strong electrostatic field. It is believed that the electric field will partly free the wax from heterogeneous parts which are usually present in the form of ions from impurities.

In conclusion the author wishes to thank Dr. L. L. Barnes and Professor E. H. Kennard for many helpful suggestions and much valuable criticism in connection with this investigation. He is further much indebted to the members of the Department of Physics, and in particular to Professor Ernest Merritt and Professor F. K. Richtmyer, who have permitted him to make use of the equipment and research room which were needed to carry out this work.

MARCH, 1935 PHYSICS VOLUME 6

An Optical Method for Measuring the Distribution of Velocity Gradients in a Two-Dimensional Flow

E. D. Alcock and C. L. Sadron, *California Institute of Technology, Pasadena, California*

(Received November 15, 1934)

I. Anisotropy of Liquids in Laminar Flow

MAXWELL discovered that some very viscous fluids (Canada balsam) when in motion present the phenomenon of double refraction. Numerous scientists have studied the phenomenon from experimental as well as theoretical points of view, the first real work being done by Kundt in 1881. The result of these investigations is that all colloidal suspensions and a large number of pure liquids show a considerable double refraction when in motion.

In the case of colloidal suspensions, theories have been advanced by several authors, not always with great success, since the experimental work shows that the effect is rather complicated. For pure liquids we have only a theory of Raman and Krishnan which is still open to discussion. Although the theory in this case is more difficult than in the case of suspensions, the experimental data indicate a simpler effect. When we are dealing with pure liquids we treat them in the following manner:

Let us consider a parallel or laminar two-dimensional flow along a solid boundary and suppose there is a constant gradient of velocity perpendicular to the boundary. Such a flow will

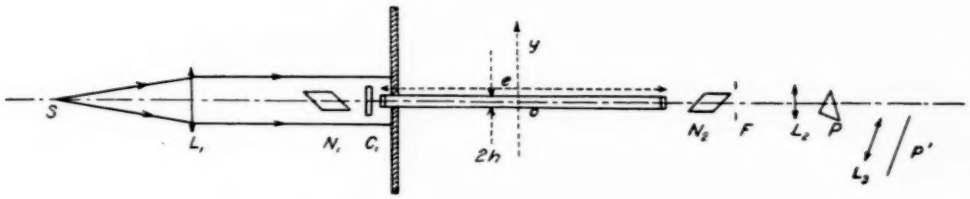


FIG. 1. Experimental arrangement. S, arc; $L_1L_2L_3$, lenses; N_1N_2, nicol prisms; C_1, gypsum wave plate; F, spectrograph slit; P, prism; P', photographic plate.

have shearing stresses parallel to the boundary. These stresses may be resolved into a tension and a compression perpendicular to each other and making an angle of 45° with the boundary. The effect of these stresses on the optical properties of the liquid is analogous to the effect produced in photoelasticity when a solid material (Bakelite, glass, etc.) is subjected to stresses. If we now send a beam of polarized light through the liquid perpendicular to the plane of the flow, the electromagnetic vector is resolved into two component vectors along the axes of tension and compression. The two components travel with different velocities due to the double refracting properties of the fluid. If the light path is of unit length, the separation of the two vibrations is given by

$$D=n_1-n_2,$$

where n_1 and n_2 are the indices of refraction in the two principle directions. The experiment (and the theory) show that the difference of the indices (n_1-n_2) is proportional to the stresses and, since a parallel flow was assumed, we have

$$D=\mu K\frac{\partial u}{\partial y},$$

where K is the so-called Maxwell constant which depends upon the molecular properties of the fluid and μ is the coefficient of viscosity.

If the velocity gradient is a function of y in a given cross section, D will be proportional to the same function of y. If our flow is curved, the double refraction at any point is given by

$$D=\mu K\left[\frac{\partial u}{\partial y}-u/R\right],$$

where R is the radius of curvature of the flow at the given point.

II. APPLICATIONS

In all cases where the radius of curvature of the flow is large so that the term u/R may be neglected, the above phenomenon furnishes us with a simple method of measuring the distribution of velocity gradients or of velocity without disturbing the flow. The problem is reduced to measuring the double refraction D and Maxwell's constant K. From these two determinations we obtain directly the velocity gradient and by a simple integration, the velocity. It is a simple problem in crystalline optics to determine the double refraction. We used a method already used by several other experimenters. The double refraction produced by the liquid is usually very small, therefore we use it to modify the double refraction of a gypsum wave plate. By measuring the shift in the interference fringe of the wave plate with a spectrometer we obtain the double refraction of the flowing liquid.

First application: Parallel channel

In order to check the value of our method we applied it to the well-known case of flow between two infinite planes. We used a channel of rectangular cross section with a very large aspect ratio. (See Fig. 1.) In this channel we circulated sesame oil with an average velocity of about 1 m/sec. by means of a pump loaned to us through the courtesy of the Byron Jackson Company. We measured this average velocity by taking motion pictures of the falling oil in the gauge and of a stopwatch simultaneously. Theoretically the flow in a cross section of this type gives us a linear distribution of velocity gradient from one wall to the other. Fig. 2 shows our experimental results, which are in quite good agreement with the theory. Because of strains in the glass side walls of our channel the fringes are

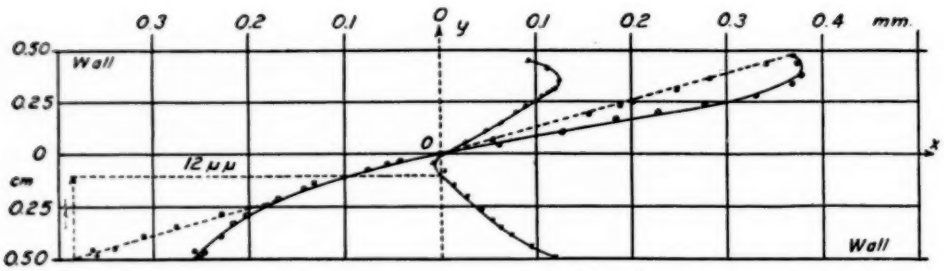


FIG. 2. Distribution of du/dy as a function of y in the section Oy.

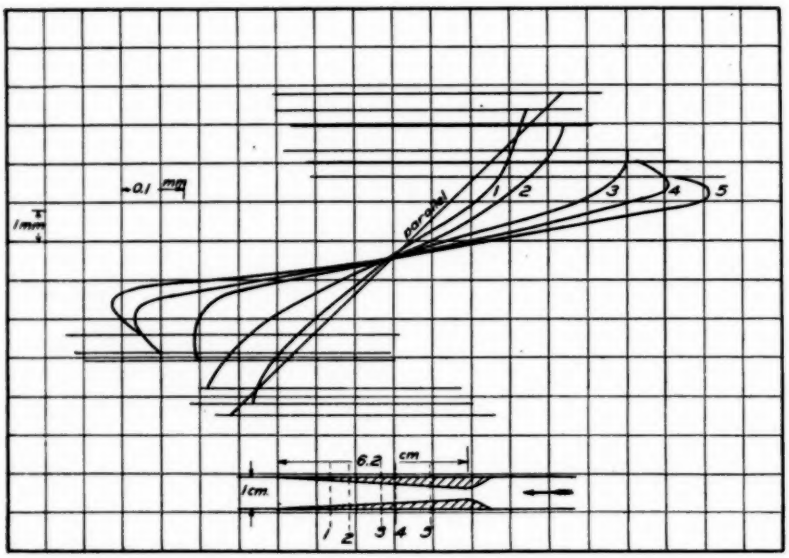


FIG. 3. Distribution of du/dy in cross section of diverging channel.

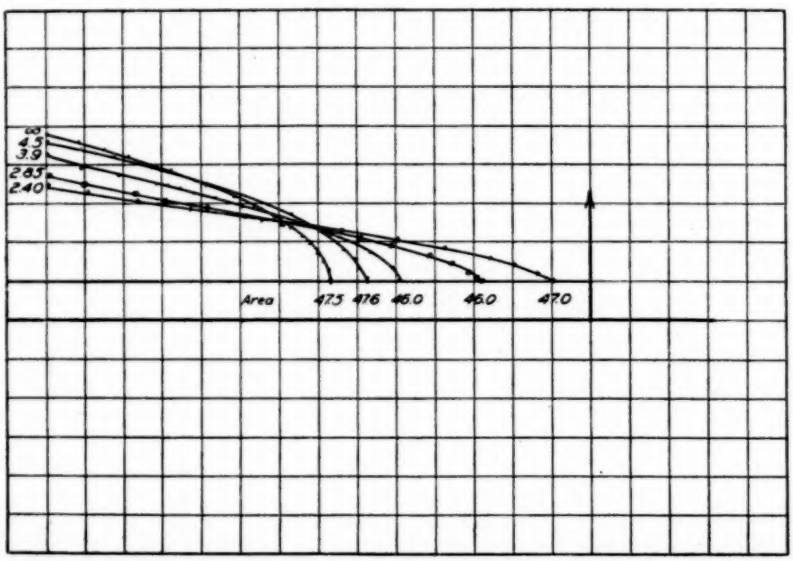


FIG. 4. Distribution of u (velocity) in cross section of diverging channel.

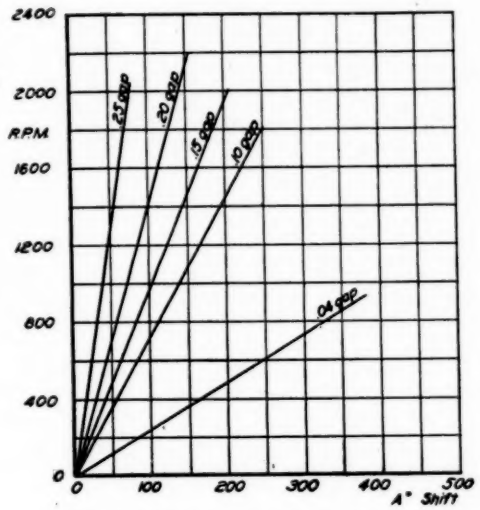


FIG. 5. Double refraction in fluid between rotating cylinders.

not straight; however, this cancels out since we only measure the shift of the fringe. By combining the theory and experiment we were able to calculate a value of Maxwell's constant of 9×10^{-11} c.g.s. Using an aspect ratio of 1/30, we computed the end effects. This calculation showed that we were introducing an error of not over 3 percent by neglecting them. The estimated accuracy of the method is about eight percent.

Second application: Diverging channel

We then applied the method to the problem of the flow in a diverging channel. The results are given in Figs. 3 and 4. Since the quantity of fluid flowing per unit time through any section is constant, the integral of the velocity across any section should also be constant. We have indicated in Fig. 4 the results of such an integration for several sections and they check very well.

Third application: Rotating cylinders

For the purpose of checking our value of Maxwell's constant obtained in our first experiment and to investigate what happens when the flow becomes turbulent, we took the case of flow between two concentric cylinders. The inner cylinder rotates while the outer remains stationary. We had several cylinders made which enabled us to measure the distributions in five different gaps. The results are shown in Fig. 5. The average value of Maxwell's constant calculated from these results is 8.5×10^{-11} c.g.s. which checks very well with that obtained in our first experiment. Using sesame oil we were not able to attain a high enough velocity to have turbulent flow. In order to have a control on the temperature of the oil we installed a water jacket on the outer cylinder.

CONCLUSIONS

We think that our method is applicable to many problems, it being subject only to the limitation that the flow be two dimensional. We see at present two main fields of application.

(1) Scientific investigation of the boundary layer. If sufficient care is taken we estimate that it is possible to measure the value of the velocity gradient to within 0.1 mm of the boundary. Therefore we can obtain the velocity distribution in this layer without disturbing the flow with any apparatus. Also since we can apply the method to curved flows where the term u/R can be neglected we can investigate several problems connected with separation.

(2) Experimental investigation of turbulent distributions. We cannot give any details at present on this problem, our work not being far enough advanced.

Finally we wish to point out that this method could be easily applied to the problem of the flow around a rotating system. Instead of using a continuous source in this case, we would have to use a stroboscopic device.

Magnetic Properties and Orientation of Ferromagnetic Particles[1]

C. W. DAVIS,[2] *U. S. Bureau of Mines, Pittsburgh, Pennsylvania*

(Received October 19, 1934)

Results of preliminary tests by direct methods show definitely that alignment of particles in a magnetic field increases their magnetization up to saturation and augments their remanence, but that coercive force is not noticeably affected.

INTRODUCTION

WHILE engaged in determining magnetic properties of mineral powders, incidental to the study of mineral physics being conducted by the metallurgical division of the U. S. Bureau of Mines,[3] the writer observed that magnetic particles of a definite size and packing density, when oriented by placing in a strong magnetic field and then demagnetized, gave higher values for remanence and induction at moderate field strengths (45 oersteds) than did undirected particles under similar conditions. After a satisfactory modified isthmus method had been developed[4] for determining magnetic properties of powders at field strengths up to about 3600 oersteds, the effect of particle orientation on these properties was studied in more detail.

EXPERIMENTAL

Natural and artificial magnetic oxides of iron, the purification and preparation of which are described elsewhere,[5] were studied.

Natural Fe_3O_4, a sample of well-crystallized magnetite from Mineville, New York, was sized between 60 and 250 mesh and found to contain 96.97 percent Fe_3O_4 (calculated from the FeO content), 0.83 percent Fe_2O_3, 1.38 percent SiO_2 and 0.57 percent TiO_2.

The artificial oxide produced by the partial gaseous reduction of hematite from Missouri was porous and impure, being a mixture of 60.07 percent Fe_3O_4 (calculated from the FeO content), 29.5 percent Fe_2O_3, 0.58 percent MgO and 7.93 percent SiO_2. It was selected because of the exceptionally high coercive force exhibited by material prepared in this way[6] and was crushed and sized between 80 and 250 mesh. The minus 250-mesh particles from each sample were discarded because it had been learned that the effect of size on the magnetic properties of powders became noticeable at smaller sizes.[7]

Magnetic measurements were made with the previously mentioned isthmus apparatus, using the point-by-point method for hysteretic determinations and taking care that all conditions except particle orientation remained constant for each pair of tests. This was accomplished by using a single sample of such a size that all of it could be packed into a sample capsule of fixed, known dimensions for both the random and regulated particle orientation; that is, the magnetic properties of the same material were determined with two different particle orientations at the same packing density and at the same field strength.

The observation of Weiss[8] that large mineral crystals had preferred direction of magnetization was extended to small crystals of magnetite in a very simple way. Controlled orientation of mineral grains was accomplished by the gradual addition of the particles while the sample capsule, placed between the poles of the magnet of the isthmus permeameter, was agitated by tapping.

[1] Published by permission of the Director, U. S. Bureau of Mines. (Not subject to copyright.)

[2] Associate chemist, U. S. Bureau of Mines.

[3] R. S. Dean, and others, Progress Reports—Metallurgical Division; 10. *Mineral Physics Studies*, Report of Investigations, 3268, U. S. Bureau of Mines (in course of publication).

[4] V. H. Gottschalk and C. W. Davis, *An Apparatus for Determining the Magnetic Constants of Mineral Powders*, Report of Investigations, 3268, U. S. Bureau of Mines (in course of publication).

[5] C. W. Davis, *Magnetic Properties of Mineral Powders and Their Significance*, Report of Investigations, 3268, U. S. Bureau of Mines (in course of publication).

[6] V. H. Gottschalk and C. W. Davis, *A Magnetic Material of High Coercive Force*, Nature **132**, 513 (1933).

[7] V. H. Gottschalk and F. S. Wartman, *Magnetization Curves for Magnetite Powders*, Report of Investigations, 3268, U. S. Bureau of Mines (in course of publication).

[8] Pierre Weiss, Thesis, Paris, 1896.

This method may or may not orient crystals in any definite crystallographic arrangement, but it must turn particles so that the direction of their resultant polarity approaches parallelism with the magnetic field in which they become oriented, and by rotation of the container, in which mineral particles of more than one axis of maximum magnetization are suspended in a liquid gelatin solution, a very definite mineral orientation should result.[9]

The use of a binder to fix the position of the particles was found to be unnecessary for our tests.

Demagnetization was effected by placing the sample within its capsule in a slowly reversing magnetic field that was gradually reduced to zero.

The differences between the magnetic properties of the samples in which the particles were aligned and those with random particle arrangement are shown by hysteresis loops.

[9] A. Goetz, A. B. Focke and A. Faessler, Phys. Rev. **39**, 169, 553 (1932); A. Goetz and A. Faessler, Phys. Rev. **40**, 1053A (1932); K. S. Krishnan, Phys. Rev. **45**, 115 (1934); A. Goetz, Phys. Rev. **45**, 282 (1934).

Results[10]

Fig. 1 shows that the magnetization of aligned particles of magnetite is considerably higher at moderate field strengths than that of random particles, the difference increasing up to about 230 oersteds and then decreasing as the field increases so that each sample approaches a $4\pi I$ value of 2850 gauss at a field strength of about 2700 oersteds. Both the percentage difference in magnetization and the difference $(\mu_1-\mu_2)$ in induction per unit field strength seem to be greatest for the two cases at the lowest field strength used, but it may well be that the maximum is really reached at 19 oersteds, the coercive force of the sample.

The percentage difference in $4\pi I$'s and the difference in μ's for reduced hematite samples increases as shown in Fig. 2 so that a maximum is reached at a field strength between 230 and 254 oersteds, a value slightly higher than the coercive force of 230.

The figures show that the coercive force is practically independent of particle orientation,

[10] Data from which the curves in Figs. 1 and 2 were prepared show clearly the results and relationships discussed.

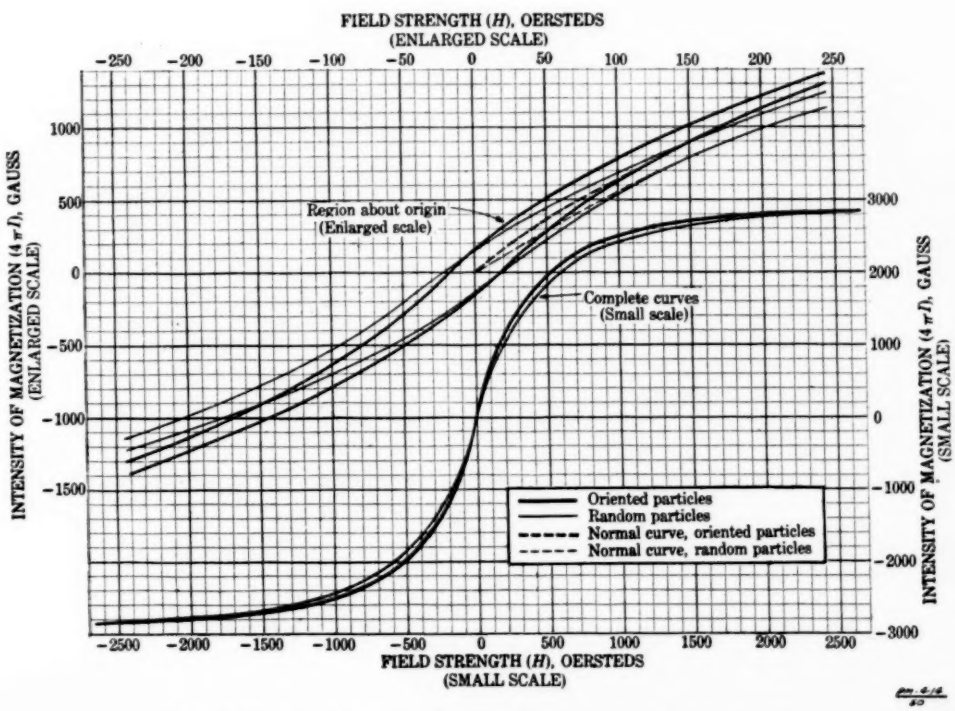


FIG. 1. Hysteresis loops and magnetization curves for magnetite.

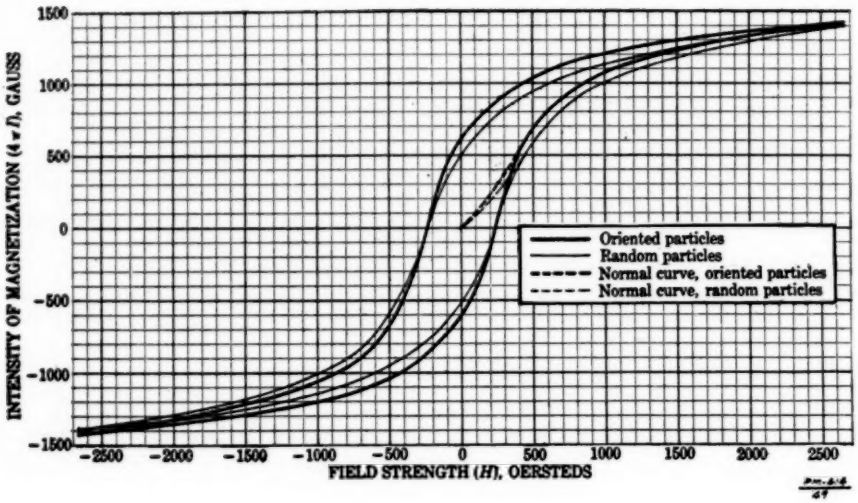


FIG. 2. Hysteresis loops and magnetization curves for reduced hematite.

the small apparent difference that may be obtained by large-scale plots being less than the probable experimental error for the determination.

The remanence, however, is seen to be substantially greater for the tests in which particles were lined up in a magnetic field, 165 as against 137 for magnetite and 610 as against 495 for partly reduced hematite.

The slight irregularities in the permeability values may or may not be accidental.

DISCUSSION

The results, which accord with the conclusions of Sizoo[11] as to remanence and induction but not as to coercive force, in general conform to the modified Weber theory which has been described by Stoner.[12] This theory, in brief, assumes that ferromagnetic substances contain a large number of domains. With fields greater than the coercive force but less than the saturation value each domain is assumed to have certain fixed directions which magnetization may follow in an attempt to approach parallelism with the impressed magnetic field. As the field is increased above saturation the directions are no longer fixed but may rotate so that parallelism to the field is then approached continuously.

The curves for the two conditions of orientation resemble those showing the "shearing effect" caused by a break in a solid substance,[13] but in our tests the samples in both states of orientation contained the same number of joints with the same distance between particles (very nearly), since the number of particles and the packing density were the same. The effect of size of particles on magnetic properties in the size range used is small.[14]

If during orientation in a magnetic field the particles turn so that the directions of the resultants of their component directions of easy magnetization are parallel to the field, it would be natural that after demagnetization the first small increment of field strength in excess of the coercive force of the mineral should cause the greatest difference in induction per unit of field strength between the controlled and randomly oriented samples. The difference approaches zero as the field increases, as would be expected if the directions of easy magnetization become aligned in the field and the two samples become more nearly magnetically alike. The magnetization does not rise to a maximum at once or even when the coercive force is reached because the individual particles contain a large number of "domains" of magnetization, but if the mag-

[11] G. J. Sizoo, *Properties of Ferromagnetic Crystals.* Physica **10**, 1–18 (1930).
[12] E. C. Stoner, *Magnetism*, pp. 61–66 (London, 1930).
[13] J. A. Ewing, *Magnetic Induction in Iron and Other Metals*, p. 279 (London, 1891).
[14] See reference 7.

netically oriented particles have a greater proportion of their "domains" in a position for easy magnetization than the random particles, the former would continue to show greater magnetization than the latter for any given field until saturation was attained.

Whether the slight irregularities of permeability exhibited by our data are due to the Barkhausen effect, to the movement of particles or to errors in the measurement of the current applied to the field magnet, has not been determined.

On reduction of field strength the hysteresis curve for the aligned lies above that for the random particles, and the remanence in the former case is considerably higher than in the latter. One important factor contributing to this difference is apparently that the magnetized particles aligned with the north end of one adjacent to the south end of the next form elongated strings which reduce the demagnetizing effect as compared to the end effect of the short unordered particles.

The fact that coercive force is independent of particle orientation indicates that the domain direction of easy magnetization is also a direction of easy demagnetization, for although the remanent magnetism may differ, the same opposed field strength effects demagnetization for different orientations.

From the theory of Dean[15] and the results of Gottschalk[16] the high coercive force of reduced hematite indicates a large interfacial area and the possibility of small ultimate particle size so that the smaller differences in magnetic properties for different orientations of the reduced hematite may be due to a more random distribution of its magnetic elements than in the crystalline magnetite. Such a condition is substantiated by the work of Doan[17] who found by x-ray diffraction methods that for minus 200-mesh particles oriented in a magnetic field the magnetite in reduced hematite was randomly oriented while Mineville magnetite was not.

[15] See reference 3.

[16] V. H. Gottschalk, *The Coercive Force of Magnetite Powders*, Report of Investigations, 3268, U. S. Bureau of Mines (in course of publication).

Concluding Remarks

Other work prevents an extension of the investigation to determine whether the study of pure crystals by the method outlined may have more fundamental significance in the field of ferromagnetism.

This method of studying the effect of particle orientation on magnetic properties offers certain advantages over those previously used. Some of the outstanding characteristics of the method are:

(1) It permits the magnetic measurement of small samples of small particles, thus enabling production and use of very pure and uniform material.

(2) It allows a single sample to be oriented in different ways without recourse to mechanical work or heat treatment, thus avoiding uncertain changes in chemical and structural composition.

(3) It permits the use of any size of particles desired without heat treatment and allows the direct microscopic measurement of the particles.

The author wishes to thank Dr. V. H. Gottschalk for his generous assistance and advice.

[17] D. J. Doan, Private communication, 1934.

Comparison of the Temperatures in a Solid and Its Scaled Model

R. V. CHURCHILL, *Department of Mathematics, University of Michigan*

(Received December 19, 1934)

If a solid is constructed of the same material and to scale with a body of any shape and subjected to initial and surface temperature conditions of the same character, properties can be derived to compare or contrast the temperatures at corresponding points in these solids. Seven properties are given in this paper, covering the cases of variable and steady temperatures of solids with and without radiation or convection at any part of the boundary, and with any fixed temperature distribution on the rest of the surface. The law used for the transfer of heat by radiation and convection is a very general one. The thermal coefficients are functions of the temperature. These properties are derived by substitutions of new variables into the heat equation and the boundary conditions. Some of the properties give important information about the effect of the size of a solid upon its rate of cooling or heating. All of the properties are useful in the determination of temperatures in any solid by measuring the temperatures in a large or small scale model.

1. INTRODUCTION

IF one body is a scaled reproduction of another the two are said to be geometrically similar. Two such solids can be placed in perspective (in an infinite number of ways) so that to each point (x, y, z) of the first there corresponds a point $(\alpha x, \alpha y, \alpha z)$ of the second, when the origin is taken as the point of similitude. The number α is positive and constant for all points, and the second solid is α times as large as the first.

When two such similar solids of the same material are subjected to the same initial and surface temperature conditions at corresponding points, their temperatures at corresponding points generally have different values at any instant. Comparisons of these temperatures are made in this paper. Considerable information is obtained about these temperatures without restricting the shape of the solid, even when the thermal coefficients are variable and the surface heat transfer law is quite general. A comparison of these temperatures in the case of steady temperatures in solids with constant thermal coefficients has already been carried out.[1]

We consider here any two geometrically similar solids of arbitrary shape, made of the same homogeneous[2] and isotropic material. The thermal conductivity can be any positive function of the temperature, $k = \kappa(T)$; likewise the specific heat and density can be functions of T. Moreover, if there is radiation (or absorption) or convection of heat, or both, at any portion of the boundary surface the flux of heat through this portion at any point can be any function of the position and temperature of this point,

$$\text{Flux} = \Phi(x, y, z, T). \tag{1}$$

This portion of the boundary will be called the "exposed surface." We define the flux as positive if heat flows into the solid through this surface.

This surface heat transfer function Φ is subjected only to the natural restriction that it cannot increase with T at any point. This is natural since it means that the absolute value of the flux cannot increase when the surface temperature is brought nearer to the temperature of the surroundings. In particular, this law of transfer includes Planck's radiation formula and the Stefan-Boltzman law of radiation, as well as the law of Dulong and Petit for radiation and convection, provided the state of the surrounding gas and radiating or absorbing bodies is kept fixed. This law also includes the condition of insulation $\Phi(x, y, z, T) = 0$ as well as that of a constant supply of heat $\Phi(x, y, z, T) = \text{constant}$, over any part of the boundary.

Any fixed temperature distribution may be assigned to the rest of the boundary surface, and the initial temperature distribution in the solid is arbitrary.

[1] R. V. Churchill, *General Properties of Steady Temperatures in Solids Partially Exposed to Gas*, Physics **4**, 50 (1933).

[2] See remarks on non-homogeneous solids in Section 5 below.

2. Comparison of Variable Temperatures in the Case of Radiation and Convection

Consider a solid of any shape with an arbitrary initial temperature distribution. Let $f(x, y, z)=0$ be the equation of that portion of its boundary surface which has a fixed temperature at each point, and let $g(x, y, z)=0$ represent the remainder of its boundary at which heat transfer by convection, radiation or absorption, or a combination of these, takes place in accordance with the general law (1).[3] The temperature T at any point at any time is then determined by the conditions,

$$c\,\partial T/\partial t=(\partial/\partial x)(k\,\partial T/\partial x)+(\partial/\partial y)(k\,\partial T/\partial y)+(\partial/\partial z)(k\,\partial T/\partial z), \tag{2}$$

$$T=T_0(x, y, z) \text{ when } t=0, \tag{3}$$

$$T=T_f(x, y, z) \text{ on } f(x, y, z)=0, \tag{4}$$

$$k\,dT/dn=\Phi(x, y, z, T) \text{ on } g(x, y, z)=0. \tag{5}$$

The heat capacity c per unit volume is any function of the temperature, and n is the outward drawn normal to the surface $g(x, y, z)=0$.

Eq. (2) can be reduced[4] to the form of the heat equation with constant conductivity by the substitution of the new dependent variable

$$u=\int_0^T \kappa(\lambda)d\lambda. \tag{6}$$

The conductivity $\kappa(T)$ is positive, so u is a monotonic increasing function of T, and Eq. (6) defines T as a unique function of u,

$$T=\tau(u). \tag{7}$$

Since $du/dT=\kappa(T)=k$, Eqs. (2)–(5) become, in terms of u,

$$\frac{\partial u}{\partial t}=(k/c)(\partial^2u/\partial x^2+\partial^2u/\partial y^2+\partial^2u/\partial z^2), \tag{8}$$

$$u=u_0(x, y, z) \text{ when } t=0, \tag{9}$$

$$u=u_f(x, y, z) \text{ on } f(x, y, z)=0, \tag{10}$$

$$du/dn=\Phi(x, y, z, \tau(u))=\phi(x, y, z, u) \text{ on } g(x, y, z)=0, \tag{11}$$

where

$$u_0(x, y, z)=\int_0^{T_0(x, y, z)}\kappa(\lambda)d\lambda,$$

$$u_f(x, y, z)=\int_0^{T_f(x, y, z)}\kappa(\lambda)d\lambda.$$

Let the solution of this boundary value problem be

$$u=U(x, y, z, t). \tag{12}$$

We now substitute new independent variables throughout this problem,

$$x'=\alpha x, \quad y'=\alpha y, \quad z'=\alpha z, \quad t'=\alpha^2 t, \tag{13}$$

where α is a positive constant. If λ, μ, ν are the direction cosines of the normal n, then

$$du/dn=\lambda\partial u/\partial x+\mu\partial u/\partial y+\nu\partial u/\partial z=\alpha(\lambda\partial u/\partial x'+\mu\partial u/\partial y'+\nu\partial u/\partial z'). \tag{14}$$

We shall call the expression in parentheses du/dn'.

When these new variables are substituted the factor α cancels in Eq. (8), and conditions (8)–(11) become

$$\partial u'/\partial t'=(k/c)(\partial^2u'/\partial x'^2+\partial^2u'/\partial y'^2+\partial^2u'/\partial z'^2) \tag{15}$$

$$u'=u_0(x'/\alpha, y'/\alpha, z'/\alpha) \text{ when } t'=0, \tag{16}$$

$$u'=u_f(x'/\alpha, y'/\alpha, z'/\alpha) \text{ on } f(x'/\alpha, y'/\alpha, z'/\alpha)=0, \tag{17}$$

$$du'/dn'=(1/\alpha)\phi(x'/\alpha, y'/\alpha, z'/\alpha, u') \text{ on } g(x'/\alpha, y'/\alpha, z'/\alpha)=0. \tag{18}$$

Here u' is the function of x', y', z', t' into which u transforms under the substitutions (13), so according to Eq. (12) the solution of this problem is

$$u'=U(x'/\alpha, y'/\alpha, z'/\alpha, t'/\alpha^2). \tag{19}$$

Now let x', y', z' be interpreted as the coordinates of a point, and t' as time, in terms of the same units of length and time as those used originally in Eqs. (8)–(12). Eqs. (15)–(19) then have a new physical interpretation.

A surface $F(x/\alpha, y/\alpha, z/\alpha)=0$ is in perspective with the surface $F(x, y, z)=0$, with the

---

[3] Either of the functions f and g may be represented by more than one analytic expression. For instance $f(x, y, z)=0$ may represent one base and the lateral surface of a cylinder.

[4] This has been pointed out by M. S. Van Dusen, *Note on the theory of heat conduction*, Bur. Standards J. Research **4**, 753 (1930).

constant α for its ratio of perspectivity, and with (x, y, z) and $(x/\alpha, y/\alpha, z/\alpha)$ as corresponding points. Hence Eqs. (17) and (18) are the surface boundary conditions for the function u in a solid in perspective with the original solid. A comparison of the conditions (17) and (10) show that the value of u' at (x', y', z') on the surface $f(x'/\alpha, y'/\alpha, z'/\alpha)=0$ is equal to $u_0(x'/\alpha, y'/\alpha, z'/\alpha)$, which is the value of u at the corresponding point on the boundary of the original solid. It is easily seen that the direction cosines are the same at corresponding points of two surfaces in perspective. So, according to Eq. (14), du/dn' is the normal derivative of the function u' at the boundary of the new solid. A comparison of Eqs. (18) and (11) shows that the normal derivatives of u' and u at corresponding points on the surfaces of the two solids vary with u according to the same law except for the factor $1/\alpha$ in Eq. (18).

According to Eqs. (16) and (9) the initial values of u' and u are the same at corresponding points of the two solids. Eqs. (15) and (8) show that u' and u satisfy the same differential equation throughout the solids. Finally, a comparison of Eqs. (19) and (12) shows that the value of u' at any point of the new solid at time t is equal to the value of u at the corresponding point of the original at time t/α^2,

$$u' = U'(x', y', z', t') = U(x'/\alpha, y'/\alpha, z'/\alpha, t'/\alpha^2).$$

Since k and c are the same functions of T in both solids, it follows from Eq. (7) that the T's are equal when the u's are equal; also if the u's are unequal the T's are unequal in the same sense. The first property is thus established.

Property I. If a solid is constructed to the scale α with another and subjected to the same initial temperature and surface temperature conditions at corresponding points, except that its surface heat transfer function is $1/\alpha$ times the function for the original, the temperature at each of its points at time t is the same as the temperature at the corresponding point of the original at time t/α^2.

A less involved property arises from this if we assume that the thermal coefficients vary so that the ratio k/c is constant. This condition is closely approximated in various applications. Under this assumption a change in the character of the surface heat transfer which causes the function ϕ to be replaced by ϕ', where

$$\phi'(x, y, z, u) \leqq \phi(x, y, z, u), \tag{20}$$

will produce a new u less than or equal to the original.

To prove this we first write the conditions which determine u', the new u:

$$du'/dt = (k/c)(\partial^2 u'/\partial x^2 + \partial^2 u'/\partial y^2 + \partial^2 u'/\partial z^2), \tag{21}$$

$$u' = u_0(x, y, z) \text{ when } t=0, \tag{22}$$

$$u' = u_f(x, y, z) \text{ on } f(x, y, z)=0, \tag{23}$$

$$du'/dn = \phi'(x, y, z, u') \text{ on } g(x, y, z)=0. \tag{24}$$

Conditions (8)–(11) determine u in the same solid. By subtracting each equation in the set (21)–(24) from the corresponding one in the set (8)–(11) and writing $w=u-u'$, we find that w must satisfy the conditions

$$\begin{aligned} &\partial w/\partial t = (k/c)(\partial^2 w/\partial x^2 + \partial^2 w/\partial y^2 + \partial^2 w/\partial z^2), \\ &w=0 \text{ when } t=0, \\ &w=0 \text{ on } f(x, y, z)=0, \\ &dw/dn = \phi(x, y, z, u) - \phi'(x, y, z, u') \\ &\qquad \text{on } g(x, y, z)=0. \end{aligned} \tag{25}$$

These conditions show that w can be interpreted as the variable temperature in the given solid with initial temperature zero and temperature of the surface $f(x, y, z)=0$ held at zero, while heat transfer takes place at the surface $g(x, y, z)=0$. However, if $w \leqq 0$ at any point of this surface then $u \leqq u'$ there and

$$\phi(x, y, z, u) \geqq \phi(x, y, z, u'),$$

since ϕ is a non-increasing function of T and therefore of u. Hence

$$\begin{aligned} &\phi(x, y, z, u) - \phi'(x, y, z, u') \\ &\qquad \geqq \phi(x, y, z, u') - \phi'(x, y, z, u') \geqq 0, \end{aligned}$$

according to the condition (20). It follows from Eq. (25) that, at each point of $g(x, y, z)=0$,

$$dw/dn \geqq 0 \text{ when } w \leqq 0. \tag{26}$$

Let us now resort to physical argument. Since n is the outward drawn normal to $g(x, y, z)=0$ the condition (26) states that heat cannot flow out of the solid at any point on this surface whose temperature w is zero or lower. Since the

initial temperature is zero throughout and the remaining surface $f(x, y, z)=0$ is held at zero temperature, it is evidently impossible for w to fall below zero at any point on the boundary. It follows from the second law of thermodynamics that $w \geqq 0$ throughout for all t. Hence when the condition (20) is satisfied, $u'(x, y, z, t) \leqq u(x, y, z, t)$ throughout.

We now apply this result to Property I. If $\phi(x, y, z, u) \geqq 0$ on $g(x, y, z)=0$, then $(1/\alpha) \times \phi(x, y, z, u) \leqq \phi(x, y, z, u)$ when $\alpha > 1$. Hence when the transfer function ϕ/α obtains, the value of u is not greater than that resulting from the transfer ϕ. A similar argument applies if $\phi(x, y, z, u) \leqq 0$, so we have proved

Property II. If a solid is constructed to the scale $\alpha > 1$ with another and subjected to the same initial and surface temperature conditions at corresponding points, the temperature at each of its points at time t is greater than or equal to the temperature at the corresponding point of the original at time t/α^2 as long as the flux of heat through the exposed surface has not been outward at any point. If this flux has not been inward then the temperature is less than or equal to the corresponding temperature at t/α^2. Here it is assumed that k/c is constant for these solids.

A less accurate statement is the following: if the size of a solid is increased by a factor α, it heats more than $1/\alpha^2$ times as fast and it cools more than $1/\alpha^2$ times as fast.

3. Comparison of Variable Temperatures in Case There Is No Radiation or Convection

We now consider the important special case in which

$$\Phi(x, y, z, T)=0 \quad \text{on} \quad g(x, y, z)=0. \tag{27}$$

As before, k and c are functions of T, the initial temperature throughout the solid and the fixed temperature on the surface $f(x, y, z)=0$ are arbitrary, but the remainder of the boundary is insulated (condition (27)). Our next property follows directly from Property I.

Property III. When part of the boundary surface is held at a fixed temperature distribution while the rest is insulated, an increase in the size of a solid by a factor α causes an increase in its heating or cooling time by a factor α^2.

More precisely, if a solid is constructed to the scale α with a given solid and subjected to the same initial and surface temperature conditions at corresponding points, the temperature at each point at time t equals the temperature at the corresponding point of the original at time t/α^2. Thus it takes this solid α^2 times as long to attain a temperature at each point equal to that at the corresponding point of the given solid.

In particular, if the temperature at any point in the given solid is always increasing, the temperature at the corresponding point in the solid α times as large at the same instant is lower if $\alpha > 1$.

Property III also applies to solids whose entire boundary is subjected to a fixed temperature distribution (condition (4)).

4. Comparison of Steady Temperatures

The steady temperature is the limit of our variable temperature T, as $t \to \infty$. If this exists at each point of the solid then the steady value $\bar{u}$ of the function u is obtained from Eq. (6),

$$\bar{u}=\lim_{t\to\infty} \int_0^T \kappa(\lambda)d\lambda.$$

By interpreting x', y', z' again as the coordinates of a point in a solid α times as large as the original, it follows from Eq. (19) that

$$\bar{u}' = \lim_{t'\to\infty} U(x'/\alpha, y'/\alpha, z'/\alpha, t'/\alpha^2)$$

$$= \lim_{t\to\infty} U(x'/\alpha, y'/\alpha, z'/\alpha, t).$$

According to Eq. (12) this second limit is the steady value of u at the corresponding point of the original solid, so $\bar{u}'=\bar{u}$ at corresponding points. If ϕ is continuous then it follows that

$$\lim_{t'\to\infty} \phi(x'/\alpha, y'/\alpha, z'/\alpha, u') = \lim_{t\to\infty} \phi(x'/\alpha, y'/\alpha, z'/\alpha, u);$$

so the steady values of the transfer functions are equal at corresponding points, except for the factor $1/\alpha$ in condition (18). Hence for solids

whose thermal coefficients are functions of temperature, we have

Property IV. When the size of a solid is increased by the factor α with surface temperature conditions kept the same at corresponding points except that the transfer function is multiplied by $1/\alpha$, the steady temperatures at corresponding points are equal.

The steady function $\bar{u}$ satisfies Laplace's equation, since $\partial\bar{u}/\partial t=0$ (Eq. (8)). But $\bar{u}$ is a monotonic increasing function of the steady temperature, so the properties of steady temperatures already derived[1] apply to $\bar{u}$ and hence to steady temperatures in solids with variable thermal coefficients. We state the two of greatest importance below.

Property V. An increase in the size of a solid by a factor α produces the same change in the steady temperature (at corresponding points) as that caused by multiplying its transfer function $\bar{\phi}$ by α.

That is, if a body is constructed of the same material and to the scale α with a given one, with the same fixed temperature at corresponding surface points and the same transfer law at all other corresponding points on the surface, its steady temperature at each point equals that which would be obtained at the corresponding point of the original by multiplying the function ϕ of the original by α.

Property VI. An increase in the size of a solid causes an increase in its steady temperature at corresponding points if the solid is absorbing heat at its exposed surface ($\bar{\phi}>0$), and a decrease if it is losing heat through this surface. The fixed temperature surfaces are excepted, of course.

In case there is no radiation or convection at the surface, $\phi(x, y, z, \bar{u})=0$ on $g(x, y, z)=0$. Property IV then becomes

Property VII. When two solids are constructed to scale with corresponding parts of their boundaries held at the same fixed temperatures and the rest insulated, the steady temperatures at corresponding points throughout are the same.

5. Discussion

These properties appear to be useful in a variety of problems of design involving large or small scale models when it is desirable to determine temperatures from these models.

Property I, the most general of the above properties, enables one to determine the temperature at any point in a solid at any time from the temperatures in a scaled model, provided the transfer of heat at the exposed surface can be properly controlled. If this control is not possible so that the same law of heat transfer at the exposed surface must be applied to the model and the original solid, then Property II can be used. This furnishes a lower limit for the temperature at each point of a solid in terms of a corresponding temperature in a small scale model, provided the solids are absorbing heat through their exposed surfaces. It furnishes an upper limit if the solids lose heat through these surfaces.

The application of the rest of the properties, dealing with insulated surfaces and steady temperatures, to determine the temperatures in a body from those in a model is more direct and quite obvious.

The condition (19) or its equivalent,

$$U'(x, y, z, t)=U(x/\alpha, y/\alpha, z/\alpha, t/\alpha^2),$$

shows that the $u'=$constant surface at time t/α^2 is in perspective with the $u=$constant surface at time t, with α as the ratio of perspectivity. Hence in a scaled model satisfying the conditions of Property I or Property III, every isotherm in the model corresponds to a geometrically similar one in the original solid; but the temperatures on these isotherms are equal at different times.

Under the conditions of Property IV or Property VII (steady temperatures) each isotherm in the model is geometrically similar to the corresponding isotherm in the original solid. The ratio of magnification is the same for the isotherms as for the solids.

For composite bodies consisting of two or more homogeneous solids in perfect thermal contact, Property I still holds true under the same general initial and boundary conditions. Property III is also true for composite solids since it is a special case of Property I. The proof will be given in a later paper in which a more complete investigation of the problem for non-homogeneous solids will be made.

Magnetic Anisotropy in Silicon Steel

K. J. SIXTUS, *General Electric Company, Schenectady, N. Y.*

(Received December 3, 1934)

In hot rolled and in cold rolled sheets of silicon steel there are directions of easiest magnetization both before and after the final high temperature anneal. They are exhibited clearly by a disk suspended in a magnetic field. This property can be explained by the fact that a great part of the crystals composing the sample lie in a preferred orientation. The samples thus exhibit to a higher or smaller degree the anisotropic magnetic properties of single crystals. This non-random orientation of the crystals was shown for the annealed samples by a determination of the orientation of a great number of crystals by a light reflection method. The preferred orientations in cold rolled and hot rolled annealed samples differ substantially.

GENERAL SURVEY

EARLY in the development of silicon steel as used in transformers and rotating electrical machinery, it was recognized that the sheets showed a nonuniformity in their magnetic qualities. With one and the same sheet, it was found that permeability and hysteresis loss had different values depending on the direction in the plane of a sheet in which they were measured. This was true both immediately after the sheets had been rolled to their final thickness and also after the following final anneal which improved the magnetic properties in general. The nonuniformity resulted in the requirement in the Epstein test for magnetic losses that half of the total material tested must be measured with the flux in the rolling direction, and half of it with the flux perpendicular to this direction. Thereby a certain average of the qualities of the material was obtained.

Recently, Dahl and Pfaffenberger[1] have employed a very simple method by which the inhomogeneous properties of magnetic samples can be demonstrated in a convincing way. A disk is punched from the sheet which is to be investigated and suspended in a magnetic field of sufficient strength. If this disk were homogeneous (isotropic) magnetically, it would remain in any position in the field. If, however, the material has a higher permeability in one direction than in another, the disk will assume a position in which this direction of high permeability is parallel to the field lines. In any other position a torque will be developed which tends to pull the disk into its equilibrium position.

We are here mainly concerned with the cause for this inhomogeneity, which, due to the advance in our knowledge of magnetism during the last 10 years, seems now to be definitely established. Several explanations can be and at various times have been advanced. The fact that the grains are elongated in the rolling direction might explain a smaller loss in that direction than perpendicular to it because there the flux has to pass fewer obstructing grain boundaries. It would, however, hardly explain the fact that in certain cases the losses are lowest at 45° to the rolling direction, and it certainly would not hold for a sheet after anneal where the grain dimensions are, on the average, the same in all directions. The same arguments rule out internal stresses as the cause for the nonuniformity, although they will probably have some effect before annealing. Messkin and Kussmann[2] ascribe the directional properties to non-metallic inclusions elongated in the rolling direction by the process of rolling which would not be affected by the annealing. But this effect, just as the first one mentioned, could not explain the presence of a lowest loss direction at 45° to the rolling direction.

The correct interpretation was made possible through our advanced familiarity with the magnetic properties of individual crystals of ferromagnetic materials. Little was known about them as long as one dealt only with aggregates of many small crystals such as are met with in commercial steel. Later, by special methods, it

[1] E. Dahl and F. Pfaffenberger, Zeits. f. Physik **71**, 93 (1931).

[2] W. S. Messkin and A. Kussmann, *Die Ferromagnetischen Legierungen*, 1932.

was possible to produce large crystals on which magnetic measurements could be performed. Ruder,[3] in 1917, made strips of Si steel of up to 10 inches in length, which in their entire length consisted of one single grain. Such single crystals, as they are commonly called, have different magnetic properties in different directions just as rolled sheets have, and a possible connection suggested itself. If it could be proved that the small crystals in rolled material were not oriented at random, but showed certain preferred orientations, then the observed anisotropy could easily be explained as being caused by the magnetic anisotropy of the individual crystal. This proof was furnished for unannealed material, where we deal with a fine grained fiber structure, by an x-ray analysis using the Debye-Scherrer-Hull method. In the case of annealed material, however, where the grain size is too large, x-ray analysis can, in general, give us no information as to whether or not a preferred crystal orientation is present. Goss[4] has recently published x-ray Laue diagrams of annealed cold rolled specimens, and has concluded without justification, in the light of our results given below, that no preferred orientation was present. An optical method in which the crystal orientation is determined by light reflections from the etched surface, and which will be described later, showed quite definitely that a preferred orientation was also present after anneal in hot and cold rolled sheets.

Magnetic Properties of Single Crystals of Iron

At this point we stop to consider the magnetic properties of single crystals, firstly to show some properties of the crystalline state, and secondly to analyze what information could be obtained from measurements with the simple torque apparatus mentioned in the beginning.

Instead of Si steel we choose pure iron whose properties are better known, but which, for our purpose, is representative of the former. In an iron crystal the iron atoms have a regular distribution—they form a so-called body centered cubic lattice. The fundamental element of this lattice is a cube whose corners and whose center are each occupied by an iron atom. (Fig. 1.) It was found by measurement that up to about $I=1000$ ($B\sim 12{,}000$ gauss) the magnetization curve practically coincided for all directions in the crystal, but that above that value the magnetization was different depending on the direction in which H was applied. It took only small fields to saturate the crystal along a cube edge ([100] axis), and increasingly higher H for saturation along the face diagonal ([110] axis) and the body diagonal ([111] axis). Accordingly the six [100] axes are called the axes of easiest magnetization. According to our present view the process of magnetization along a [110] axis, for example, takes place in the following manner. The magnetization vectors in the various elementary districts of which the crystal is built up gradually, upon increase of H, grow in the two [100] directions closest to the field direction until half of all the districts are magnetized in each of these two directions, and the component of **I** in field direction is $I_s/\sqrt{2}$. This process requires practically no energy, but with further increase of H the **I** vectors swing gradually into the field direction, and the energy required is much greater. This explains the bend in the magnetization curve for the [110] axis at $I=I_s/\sqrt{2}$. The magnetization along the [111] axis takes place in a similar way, the bend appearing at $I=I_s/\sqrt{3}$. The so-called technical saturation I_s is reached when **I** lies parallel to H.

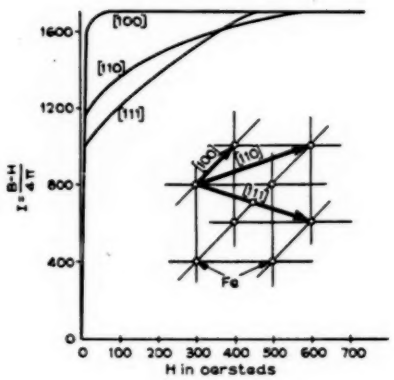


FIG. 1. The magnetization curves of iron along three crystal axes which are indicated on the elementary cube shown as insert.

[3] W. E. Ruder, Trans. Am. Soc. Steel Treating **8**, 23 (1925).
[4] N. P. Goss, Trans. Am. Soc. Steel Treating, Fall Meeting, 1934.

It is quite evident from these facts that a single crystal disk, having different permeabilities in the different crystal directions, would orient itself in a magnetic field which is sufficiently strong to produce an induction above the first bend. The same follows logically for a polycrystal whose crystals are not oriented at random, but show certain preferred orientations. The mathematical expression for this torque can be derived easily under certain simple conditions.

Akulov[5] first set up the fundamental energy equation for ferromagnetic crystals, free of strains, expressing their free energy F in terms of the direction which magnetization **I** and field H have in the lattice.

$$F=F_0(I)+2k(\alpha_1^2\alpha_2^2+\alpha_2^2\alpha_3^2+\alpha_3^2\alpha_1^2) -HI_s(\alpha_1\beta_1+\alpha_2\beta_2+\alpha_3\beta_3). \quad (1)$$

$F_0(I)$ is the energy necessary to magnetize the crystal along a 100 axis up to the intensity I, k is a constant, α_1, α_2, α_3 are the direction cosines of the magnetization **I** with respect to the three main crystal axes (I_x/I_s, $\cdots$) and β_1, β_2, β_3 the direction cosines of H with these axes (H_x/H, $\cdots$). Because of the second term on the right side of the equation the direction of **I** is in general not the same as that of H. In a disk, cut from this crystal and suspended in a field, this deviation will result in a torque, and if we take the particular case of a disk cut parallel to a (100) plane, we can derive the torque T from Eq. (1) by differentiation:

$$dF/d\varphi=T=k\sin 4\varphi(=HI_n). \quad (2)$$

φ is the angle between magnetization and a [100] crystal axis, I_n the component of **I** normal to H. The constant k may be derived by measuring H and I_n or T.[6] We can easily see that in a polycrystal, a certain percentage of whose crystals had a preferred orientation with a (100) plane in the surface and the rest of whose crystals were distributed at random, we would obtain a torque curve of the type of Eq. (2), only with a different constant k'. k'/k would then represent the percentage of crystals in the preferred orientation. N. Akulov and N. Brüchatov[7] have used this method for the analysis of rolled sheets and derive the percentage of crystals in certain preferred orientations without, however, offering a check by some other method. In general this way of analyzing is only possible if the direction or directions of preferred orientation are known from the start, and if the spread about this orientation is small. Thus it becomes necessary, before a quantitative interpretation of the torque curves can be undertaken, to establish the direction and degree of preferred orientation by an x-ray method, or, if that is not possible, by a light reflection method. Although a quantitative interpretation of the torque measurements thus necessitates the taking of additional data, the torque method in itself is an excellent and extremely simple means of obtaining qualitative information about the magnetic merits of a material. The greater the torque maximum, the larger will be the number of crystals that have cube edges ([100] axes) in a uniform direction. The more crystals with cube edges there are in this direction, the higher will be the permeability, again in this direction, at high fields. (See Fig. 1.) The angle between this preferred direction and the rolling direction can be read from the torque curve as the equilibrium position next to the torque maximum under consideration.

It seems advisable at this place to discuss briefly the advantages which a material with preferred orientation has over materials with random orientation, especially since a fine grain material with uniform properties in all directions was in the past considered as a condition which, although it could not be achieved, should at least be approached. The most important properties of silicon steel as used, for instance, for transformers are its permeability and its hysteresis loss. For the former one can make definite predictions on the basis of Fig. 1. Below a magnetization of $I\sim1000$ ($B\sim12{,}000$ gauss) the permeability is practically the same in all crystal directions and only little can be gained by having a preferred orientation. Above this value, for $B>12{,}000$ gauss, the increase in permeability through preferred orientation—if

[5] N. Akulov, Zeits. f. Physik **69**, 78 (1931).

[6] The torque due to rotational hysteresis is negligible at the high field strengths used.

[7] N. Akulov and N. Brüchatov, Ann. d. Physik **15**, 741 (1932).

the field is always in the direction of this preferred orientation—is very marked and a given B-value can be reached with much smaller H than without a preferred orientation. Little can be said about the effect of preferred orientation on hysteresis. In a series of measurements on single crystals a dependence of hysteresis on crystal orientation was found[8] with a minimum along the [100] axis, which would indicate that a preferred orientation might result in a reduction of hysteresis. The effect of preferred orientation on eddy current losses can reasonably be expected to be very slight.

Torque Measurements

Fig. 2 shows the apparatus developed for determining the torque at any angle between field and rolling direction. A scale is attached to the disk to be measured and when the movable pointer P is turned, it will indicate the torque in degrees at any position of the disk which in turn is observed at the stationary pointer A.

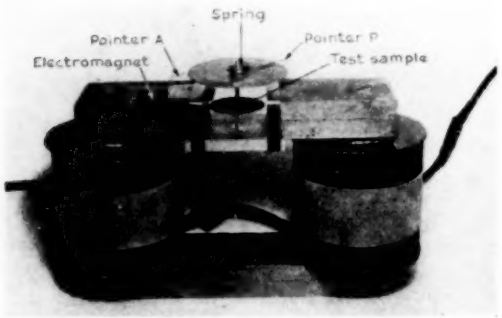


Fig. 2. Apparatus for measuring the torque exerted on a disk in a magnetic field (apparatus in an early stage of development).

By calibrating the spring the torque can be expressed in gcm or ergs instead of in degrees. The field value chosen had such a value that on further increase of the field the torque curve remained the same. Fig. 3 shows results obtained in two disks with different treatment. In a hot rolled sheet[9] there are two preferred directions at approximately $\pm 45°$ to the rolling direction both before and after annealing. In a cold rolled

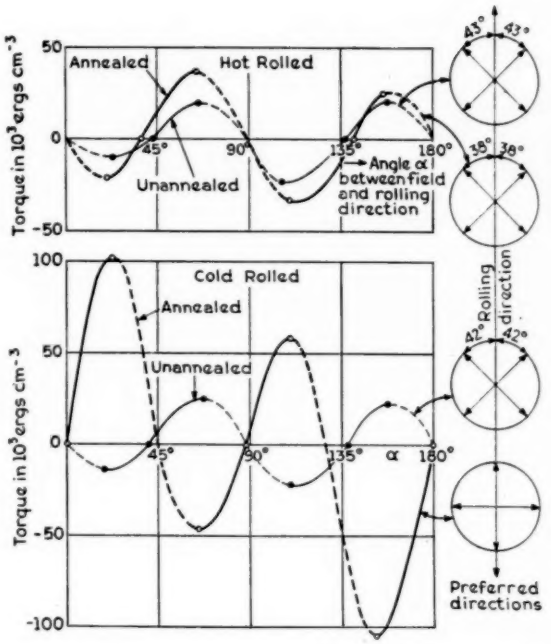


Fig. 3. Torque curves at high fields measured with the apparatus shown in Fig. 2. 3a gives the curves for a hot rolled sheet, 3b for a cold rolled sheet, both before and after anneal. H = 900 oersteds.

sheet with 85 percent cold reduction,[10] on the other hand, a distinct change takes place at the anneal. Before anneal the preferred direction is similar to the one found in hot rolled sheets, but after the anneal at 1100°C the recrystallized sheets have preferred directions which are parallel and perpendicular to the rolling direction.

This result, which shows the typical difference between hot and cold rolling found in practically all the samples investigated, could be expected from x-ray measurements as far as the preanneal sheets were concerned. In these sheets x-rays reveal a preferred crystal orientation with a [100] axis at 45° with respect to the rolling direction. For the annealed samples a light reflection method was used to indicate the preferred orientation.

Light Reflection Method

The sample surface is polished very carefully and etched for about 20 minutes in a 3 percent solution of ammonium persulfate. If the sample surface after etching is viewed in parallel light,

[8] W. E. Ruder, Trans. Am. Soc. for Metals **22**, 1120 (1934).

[9] The silicon steel used in all these experiments contained about 3.5 percent Si and 0.05 percent C.

[10] See U. S. Patent 1915766, Smith *et al.*

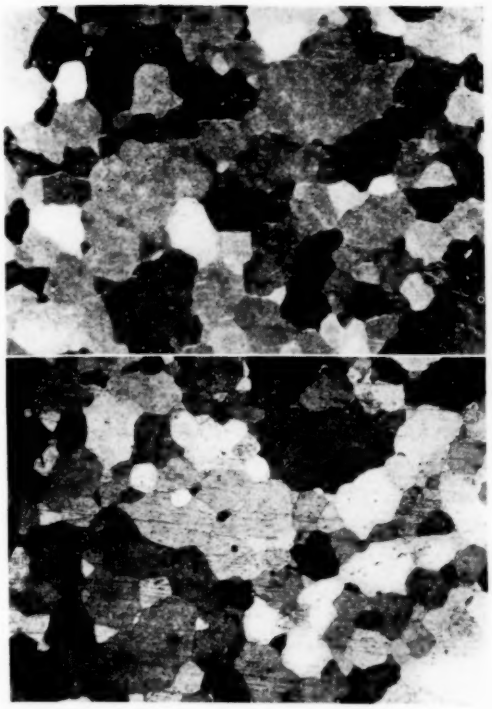
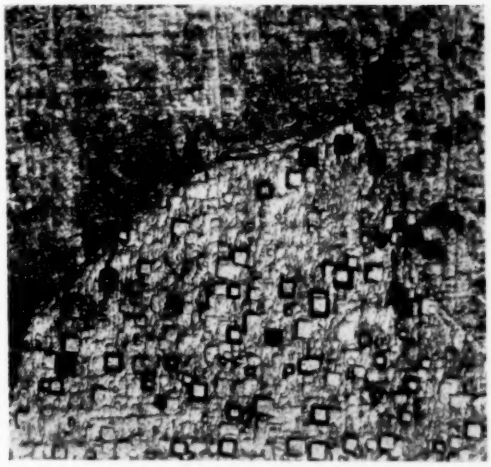


FIG. 4. Etched surface of a sample of cold rolled Si steel (20× magnification). 4a, vertical illumination; 4b, the same area with horizontal illumination with the light beam perpendicular to the rolling direction.

a number of crystals appear bright while others are dull. This is due to the peculiar way in which the crystals are attacked by the etching agent. The latter opens its attack on a crystal in a great number of places digging pits into the surface[11] whose walls (and this is the important feature) are nearly plane and parallel to cube faces, i.e., (100) planes. If we illuminate an etched sample *vertically* under the microscope all the crystals which have (100) planes parallel to the surface accordingly will appear bright (=group I), the rest dull (Fig. 4a). Group I appears dark with *horizontal* illumination and stays dark as the sample is rotated through 360°. The rest (=group II), except a few that stay dark with both vertical and horizontal illumination, go through a cycle of brightness followed by darkness during a 360° rotation, most of them twice, few of them only once (Fig. 4b). The group II crystals have very nearly (110) plane parallel to the sample surface, and, at the position of maximum brightness, have a [100] axis very nearly perpendicular to the incident light if the two brightness maxima are close to 180° apart from each other.

[11] G. Tammann, *Lehrbuch der Metallkunde*, p. 129 (1932).



→rolling direction

FIG. 5. Three crystals on the same sample as shown in Fig. 4 under higher magnification (170×).

We still do not know the orientation of the crystals with a (100) plane in the surface. This can be found with a microscope using a high magnification with which the orientation of the individual etching pits may be observed. These pits, Fig. 5, form almost perfect squares on which the direction of a [100] axis can be directly measured.

Any preferred crystal orientation can thus be discovered by determining the orientation of each individual crystal among a sufficiently large number on the specimen provided the crystals are fairly uniform in size. Such counts covering an area with about 250 crystals on each sample have been carried out by Mr. W. C. Johnson in this laboratory, and two representative determinations are plotted in Figs. 6 and 7 referring to the same samples which were used for Fig. 3. Incidentally, these counts have not only been made on the surface, but also in inner layers of the sheet after etching it to about half thickness. There, with slight variations, the same distribution curve was found. All the crystals referred to as (100) crystals reflected light vertically and had a (100) plane parallel to the surface with a spread of about ±10°. All crystals reflecting nearly horizontal light into the microscope are

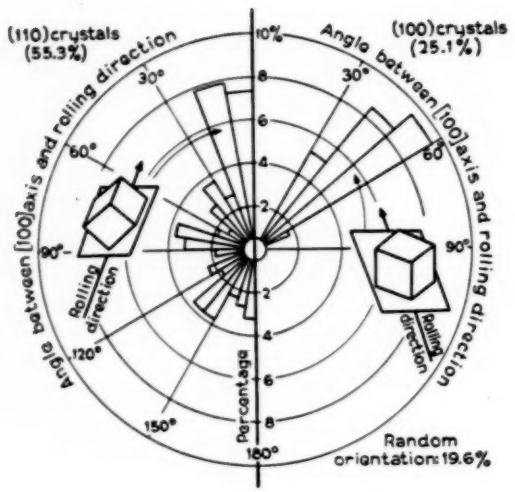


FIG. 6. The distribution of [100] axes in (100) crystals (right) and in (110) crystals (left) on the hot rolled, annealed sample of Fig. 3a. The radius of each 10° sector gives the percentage of crystals having a [100] axis within these 10°. The inserts show the preferred orientations.

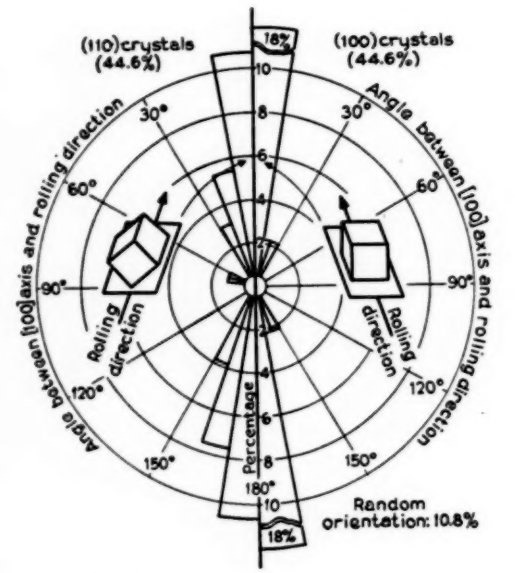


FIG. 7. The distribution of [100] axes in (100) and (110) crystals on the cold rolled annealed sample of Fig. 3b.

classified as (110) crystals if they showed two reflections at nearly 180° on rotation through 360°. The deviation in this case, however, is greater, due to greater irregularities in the etching pits, and the (110) plane in a (110) crystal may actually make an angle of up to ±20° with the sample surface. This procedure of classifying together crystals of rather diverse orientations is justifiable in our case since all of them have a [100] axis in or nearly in the surface, which is the essential property as regards magnetic behavior. All the crystals with only one and those with two reflection maxima which enclosed an angle <160° during a 360° rotation and the crystals remaining dark with both vertical and horizontal illumination are counted in the "random" class. In the hot rolled, annealed sample the (100) crystals have a decided preferred orientation with a [100] axis at ±42° to the rolling direction, whereas the (110) crystals show a slightly preferred orientation with the [100] axis in the rolling direction. In the cold rolled and annealed sample both the (100) and (110) crystals prefer a [100] axis in rolling direction. This sample has a particularly great uniformity in crystal orientation on account of an intermediate anneal.[4]

Discussion of Results

These results are significant in two respects. From a metallurgical viewpoint they prove definitely that in iron with 3.5 percent silicon, after a high temperature annealing, the crystals are not oriented at random but show a preferred orientation whose direction and degree of perfection depend on the preceding treatment. Hot rolling and cold rolling thus produce a strikingly different effect. Although their preferred orientation is nearly the same immediately after the rolling process, the orientation in the cold rolled sheet—probably due to effects on slip planes and to strains—is entirely different from the hot rolled sample after recrystallization. Secondly, from a magnetic viewpoint, the findings agree qualitatively with the torque measurements and they also explain readily some of the magnetic properties. In hot rolled sheets, e.g., among the three directions at 0°, 45° and 90° with respect to the rolling direction, the highest permeability is found at 45°, with the 0° and 90° direction as second and third best. In cold rolled sheets the 0° direction is superior to the 90° and the 45° one, and in both cases the preferred orientation among the (100) crystals is the cause.

Our findings enable us to test the accuracy of the method proposed by Akulov and Brüchatov[7] for deriving the percentage of crystals in the

preferred orientations from a torque curve. This was done by using the equations given in their paper which are derived from the fundamental Eq. (1), with the constant $k=1.5\times10^5$ ergs cm^{-3}. This value was determined by torque measurements on a single crystal disk of silicon steel. The results are given in Table I with the per-

TABLE I.

In surface → In rolling direction→	(100) plane [100] axis	[110] axis	(110) plane [100] axis
Hot rolled sample	none	16(21)	29(23)
Cold rolled sample	0(40)	none	66(35)

centages derived from our crystal count added in parentheses.

The agreement for the hot rolled specimen is satisfactory, whereas no check is obtained in the case of the cold rolled sample. The latter result shows clearly that two different crystal distributions may result in the same torque curve. It is difficult to understand this possibility at first. If, however, one takes account of the possible spread of the (100) and (110) planes given above, the two preferred orientations given in Fig. 7 are very nearly equivalent to one with a [100] axis in rolling direction and random distribution of the other [100] axes, and the result thus becomes more plausible. The two examples show that a quantitative interpretation of the torque curves is not possible in general.

It was the main object of this paper to describe the two methods used for the investigation of magnetic directional properties. In a following article quantitative results on materials after different treatments will be given.

In conclusion the author wants to express his thanks to Mr. W. E. Ruder for many discussions on the subject and to Mr. W. C. Johnson for making the tests.

MARCH, 1935 PHYSICS VOLUME 6

Growth of Crystals of Zinc Containing Cadmium by the Czochralski-Gomperz Method

HAROLD K. SCHILLING, *Physical Laboratory, University of Iowa*

(Received November 19, 1934)

In previous experiments it was found impossible to duplicate results of Hoyem and Tyndall who found a "range" for the successful growth of single crystals of zinc. Most specimens were "optically mosaic." It is now shown that by adding Cd to Zn of highest purity, single crystals may be grown and that a "range" is connected with their growth. The range is similar to, but not identical with, that of Hoyem and Tyndall. Moreover, the shape of the range is dependent on the Cd content and on the speed of growth of the crystals. Optical mosaics as well as the changes of Hoyem and Tyndall are found outside of the range. In general optical mosaics are less frequent the higher the Cd content. Certain surface markings appear on the crystals even for only 0.20 percent Cd. These are believed to be identical with markings previously described by Rosbaud and Schmid and by Straumanis and probably indicate concentration of Cd along basal planes.

INTRODUCTION

INVESTIGATIONS of the conditions governing the growth of zinc single crystals have been made in this laboratory by Hoyem and Tyndall,[1] Schilling[2] and Cinnamon.[3] The Czochralski-Gomperz method was used for the first two investigations and a modified Kapitza method for the last. Hoyem and Tyndall found that single crystals of zinc, 2.7 mm in diameter, 10 cm long, whose orientations were predetermined by "inoculation" with a nucleus, could be grown at the speed of 1.2 cm/min. only under very restricted temperature conditions. It was necessary for them to keep the temperature of the molten zinc between upper and lower limits, only 4°C apart for orientations between 0° and 55° but widening out rapidly for higher orienta-

[1] A. G. Hoyem and E. P. T. Tyndall, Phys. Rev. **33**, 81 (1929).
[2] H. K. Schilling, Physics **5**, 1 (1934).
[3] C. A. Cinnamon, R. S. I. **5**, 187 (1934); W. J. Poppy has recently checked and extended Cinnamon's results, Phys. Rev. **46**, 815 (1934).

tions. Outside this region[4] of successful growth a crystal which started successfully[5] changed to one of different orientation. They concluded that the determining factor in the successful growth of a crystal of a given orientation was a suitable temperature gradient in the liquid column below the growing crystal.

Later Hoyem grew within the H–T range a large number of successful crystals of "Kahlbaum" zinc.[6] He also found that outside the range failures (change to a crystal of different orientation) resulted. Immediately afterwards he also grew crystals of "spectroscopically pure" zinc within the same range.[7] It seemed desirable to investigate this matter further for other speeds of growth, sizes of crystals, etc. From the "successful ranges" which were expected to be found it was hoped to throw more light on the process of crystal formation. In attempting this, however, the writer found himself quite unable to reproduce the H–T results. Thinking that differences in the nature and amounts of impurities in various brands of zinc might well influence the conditions of crystal growth, altogether nine brands and lots of zinc ranging from C.P. grade to grades certainly less than 0.01 percent impure,[8] were used—all with the same results. Instead of single crystals optical mosaics[2] were obtained mainly. There were no "sudden" or "gradual" changes and therefore no range whatever. No changes in technique, nor use of other speeds of growth[2] altered the general character of the results. Further, two independent observers, Professor E. P. T. Tyndall and Dr. A. W. Hanson, kindly checked this conclusion using some of the writer's material.

Meanwhile, Poppy[3] and Hanson,[9] using a different method, had obtained definitely non-mosaic crystals of lots of zinc with slightly less than 0.01 percent impurity, and Cinnamon[3] had even established for successful growth of single crystals of the same sort of zinc, conditions very similar to the H–T range. All these workers found, however, that optical mosaics were likely to occur for very pure zinc and that special precautions[3] were then necessary to prevent mosaic starts before single crystals could be grown.

As the writer's zincs were not all of high purity, it was not possible to conclude that optical mosaics were characteristic solely of very pure zinc, nor could it be supposed that the H–T range and the production of single crystals were simply characteristic of impure zinc, because it should then have appeared for some of the writer's zincs. It seemed necessary, rather, to suspect it of being characteristic of some one particular impurity which might have been absent from all the writer's zincs but present in the H–T zincs and later in the Kahlbaum zinc of Hoyem. Comparison by spectroscopic analysis[10] showed definitely that all the zincs which had by the C–G method yielded mosaics, had less (in many cases far less) Cd impurity than the lots for which the H–T range was established and the Kahlbaum zinc later used by Hoyem.

This led to the hypothesis that the mosaics might be characteristic of zinc containing no (or *very* little) Cd, but that by adding Cd to such material single crystals might be grown by the C–G method and that the H–T range might even be duplicated for some particular percentage of Cd. Preliminary experiments by Professor Tyndall showed that by adding two or three-tenths of one percent of Cd to very pure zinc[11] the mosaics were indeed largely prevented, that the "sudden" changes returned and that signs of a range of successful growth again appeared. This paper reports subsequent investigation of the matter for various additions of Cd to Zn.

[4] This will hereafter be called the H–T range.

[5] Crystals were considered to have started growth successfully if they grew single and with the predetermined orientation for at least 1 cm. The chief enemy to a successful start was the failure to get a good junction between the nucleus and the new crystal due to inclusion of oxide.

[6] A. G. Hoyem, Phys. Rev. **38**, 1358 (1931). That these crystals were certainly good has been shown by later evidence. See Poppy, reference 3.

[7] These were considered single at the time but were under some suspicion on account of their anomalous electrical resistivities. See E. P. T. Tyndall and A. G. Hoyem, Phys. Rev. **38**, 820 (1931). Lately reexamination has shown that some of these certainly were mosaic. See Poppy, reference 3. These S. P. crystals and the Kahlbaum crystals immediately preceding them were grown in a non-oxidizing atmosphere, a fact which must be taken into account in making comparison between their growth and the growth of crystals in air, as in the present work.

[8] In a previous publication, reference 2, the maximum impurity was stated erroneously to be 0.001 percent. The above statement gives the correct figure.

[9] A. W. Hanson, Phys. Rev. **44**, 320 (1933).

[10] Carried out by Professor E. P. T. Tyndall.

[11] Horsehead-Special zinc produced by the New Jersey Zinc Company; also used by the writer in all of the present work.

Apparatus and Procedure

The apparatus was the same as that used during the writer's earlier work.[2] The growing procedure was essentially that of Hoyem and Tyndall. However, instead of using a stream of CO_2 to keep the surface of the molten zinc free of oxide, a speck of solid $ZnCl_2$[12] was dropped onto the surface just before inserting the nucleus. This melts and dissolves the surface oxide, assuring a good junction between the nucleus and new crystal. The deliberate forming of a neck[13] at the start of a crystal was found unnecessary, and the writer began drawing out the crystal as soon as the desired temperature of growth was reached. The crystals were between 2.5 and 3.3 mm in diameter and at least 10 cm long when grown at the speed of 1.2 cm/min. and 8 cm long at the speed of 0.55 cm/min.

Results and Discussion

The results obtained with 0.26 percent Cd in zinc at the speed of growth of 1.2 cm/min. (the H–T speed) are shown in Fig. 1, abscissas being orientations and ordinates $t-t_0$, where t is the temperature in degrees C of the zinc directly below the liquid column under the growing crystal and t_0 is the melting point of zinc. The black circles refer to single crystals. "Boundary" curves have been drawn in such a way that all these points, with one exception, are either definitely inside them or within 1° of them.[14] The open circles, crosses and ×'s refer to failures of three types to be described below. All of these, with only two exceptions, lie outside the region containing the successful crystals or within 1° of the nearest boundary. Hence, a range of successful growth certainly exists. Single crystals of zinc plus 0.26 percent Cd can be grown inside it with complete success while outside only failures result.

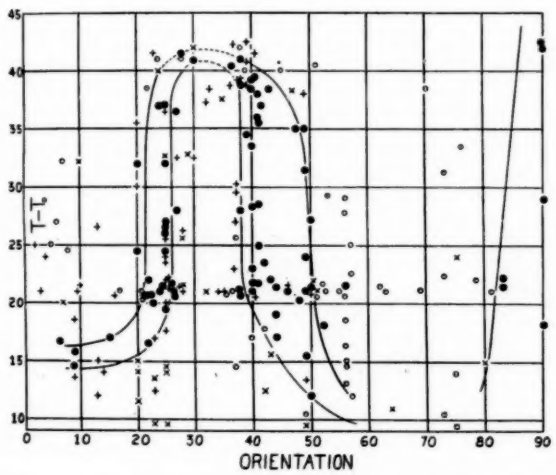


Fig. 1. Range obtained with 0.26 percent Cd+Zn at speed of 1.2 cm/min. ● single crystals; ○ failures by change from single to single of different orientation; + mosaics with no change of orientation; × mosaics with change of orientation.

Table I. *Classification of specimens of Fig. 1.*

Group	Symbol in Fig. 1	Type	Near junction	Below junction	Later change	No.
A	●	*a*	single	single	none	49
		b	mosaic	single	none	19
		c	mosaic	improving mosaic	becomes single	7
B	○	*a*	single	single	discontinuous	38
		b	mosaic	single	discontinuous	3
		c	single	single	slowly contin.	2
		d	mosaic	single	slowly contin.	0
		e	single	single	rapidly contin.	7
		f	mosaic	single	rapidly contin.	0
C	+	*a*	single	mosaic	none	17
		b	mosaic	mosaic	none	35
D	×	*a*	single	mosaic	discontinuous	5
		b	mosaic	mosaic	discontinuous	16
		c	single	mosaic	typical evolut.	5
		d	mosaic	mosaic	typical evolut.	1

In Table I is given a classification of the specimens and the number of each type obtained. The successful crystals are placed in one main group *A* and the failures in three as follows: *B*, failure by change from single crystal of one orientation to single crystal of another orientation; *C*, failure by being mosaic with no change of orientation; *D*, failure by change from mosaic of one orientation to mosaic of another. The

[12] This was first suggested by W. Fahrenhorst (see E. Schmid and M. A. Valouch, Zeits. f. Physik **75**, 532 (1932), who, however, used $ZnCl_2$ solution.

[13] See reference 1, Fig. 1.

[14] Because the crystals are very soft it is not easy to cleave them without some distortion. This makes it very difficult to determine the orientation closer than within 1°.

subdivisions of these groups are based on the character of the specimen (1) at the junction between nucleus and new crystal, (2) in the following section two or three cm in length, (3) in the rest of the specimen. Thus type *Aa* is single at the junction and remains so throughout; type *Ab* is mosaic at the junction, then becomes and remains single; *Bb* is mosaic in the junction, becomes definitely single and then changes "discontinuously"; etc. For a crystal to be mosaic at the junction means that the part of the original nucleus (always a single crystal) which is melted and then resolidified turns to mosaic (of the same orientation). Thus the new crystal gets its start as a mosaic.[15]

The types of change which may occur during growth are as follows: (1) "discontinuous," a new crystal of different orientation starts and gradually crowds out the original orientation, the specimen growing as a double crystal for several cm; this was called a "sudden" change by Hoyem and Tyndall; (2) "slowly continuous," all cleavage planes within the region of change (at least several cm) are apparently perfect, the orientation changing progressively by imperceptible amounts without the invasion of the crystal by a foreign orientation, the "gradual" change of Hoyem and Tyndall; (3) "rapidly continuous," apparently the same as the preceding type but occurring within 1 or 2 mm; (4) "typical evolution," a type of mosaic change previously described by the writer.[2]

Of the 75 group *A* crystals 68 were either single from the very start or became single very soon thereafter. Both these types (*Aa* and *Ab*) were distributed in the range quite at random. There were seven specimens (*Ac*) which remained mosaic for several cm below the start becoming gradually less mosaic (i.e., showing fewer and larger mosaic areas), finally becoming and remaining single thereafter. These were all grown very near the range boundary.

The appearance of the mosaic in the junction seems not to be connected in any way with the range, the percentages of mosaic junctions inside and outside being about equal. The phenomenon probably depends upon the precise conditions existing between the time of insertion of the nucleus and the actual beginning of growth.

Of the 129 failures (classes *B*, *C*, *D*) 55 started as mosaics but only 3 subsequently became single. Thus while all of the mosaic starts became single inside the range, practically none did outside. To start mosaic outside the range means to stay mosaic. Moreover, of the 74 failures which started single outside the range 27 (36 percent) became mosaic thereafter.

Fig. 1 shows that outside the range the region $\theta > 50°$ contains almost exclusively only group *B* failures, and that the region $\theta < 50°$ has relatively few. In the latter region group *C* and *D* specimens seem to be distributed fairly uniformly.

The directions and magnitudes of the changes of orientation are also worth noticing. For $\theta > 50°$ all changes are either toward lower orientations, or else to $\theta = 90°$ or very near it. For $\theta < 50°$ all changes are toward higher values of θ and are either small (less than about 20°), or else all the way to 90° (i.e., for discontinuous changes only).

It will be recognized at once that the writer's range resembles the H–T range in general character though considerably distorted. Both ranges are narrow, have upper and lower temperature limits with maxima for an orientation near 35° and extend upward indefinitely for θ near 90°. While both ranges determine regions of successful growth, the types of change of orientation or failures are more varied and complex in the present work.[16] In two respects, however, the present results are very different. First, there are three regions or sets of orientations (in the neighborhood of 22°, 45° and 90°) in which successful crystals can be grown over a very wide range of temperatures. Second, it appears impossible to grow crystals having orientations between about 50° and 85°, whereas one of the

[15] Compare with work of Cinnamon and Poppy, reference 3.

[16] It was early suspected that perhaps H–T had also grown mosaics but had not detected them and had counted them as singles. As examples of their "failures" some 15 short sections of specimens in which a sudden change had occurred were preserved. Recent examination of these showed that 7 were of the type *Ba* while the other 8 were good singles changing to mosaic just before a discontinuous change, i.e., these latter do not belong exactly to any of the writer's classes. An insufficient number of successful crystals were preserved to warrant the drawing of any definite conclusion. However, of nine such crystals or pieces of crystals, only two showed any indication of optically mosaic structure and both of these were grown near the border of the range.

most outstanding features of the H–T range is the wide latitude of growth conditions for single crystals having orientations above about 65°.

Experiments have also been done at the same speed of growth (1.2 cm/min.) with other concentrations (0.00 percent, 0.08 percent, 0.20 percent, 38 percent, 53 percent, 0.99 percent Cd) though the whole region of orientations and temperatures was not completely explored in each case. Throughout this work only truly single crystals containing at least 0.20 percent Cd were used as nuclei.[17] For pure zinc only type *Cb* specimens were obtained (mosaics with no perceptible change of orientation) over widely different temperature conditions. Thus we have for pure zinc a sort of stability of growth of mosaics. After adding 0.08 percent Cd, a few single crystals were obtained at low temperatures (about $t-t_0=13°$). All other specimens were of type *Dc* or *Dd*. There was no indication of a range. The results for 0.20 percent and 0.38 percent Cd were in the main the same as for 0.26 percent Cd, though the ranges obtained were narrower. For instance, in the case of 0.38 percent the range was only about half as wide at $\theta=42°$ as for 0.26 percent Cd. Apparently the Cd concentration yielding the maximum width of the range is somewhere near 0.26 percent. For 0.53 percent Cd the range was confined to a narrow strip near 90° orientation since crystals successfully started at other orientations changed soon (type *Ba*) to singles of nearly 90° orientation. It was found furthermore that with increase of Cd content (from 0.08 percent up) mosaics and mosaic changes became less frequent and never occurred for 0.99 percent Cd plus Zn. Continuous and discontinuous changes from single crystal of one orientation to single of another become, on the other hand, more frequent (outside the range, of course). With 0.99 percent Cd singles could not even be started.[18] Several crystals of different orientations always started at the junction. These went through all sorts of changes of orientations, both continuous and discontinuous, to higher and lower orientations, rapidly and slowly.

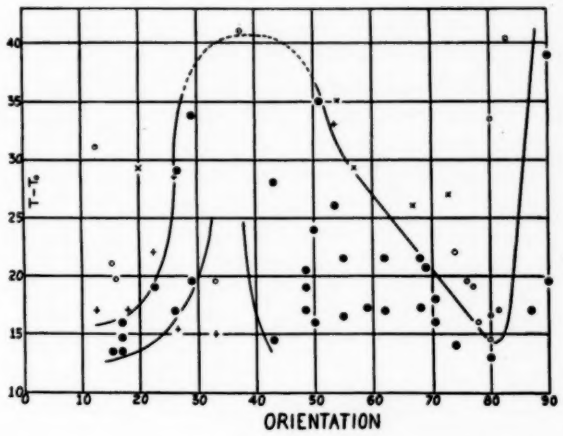


FIG. 2. Range obtained with 0.26 percent Cd+Zn at speed of 0.55 cm/min. Symbols have the same meaning as in Fig. 1.

These findings for 0.99 percent Cd in Zn seem to agree with those of Rosbaud and Schmid[19] who also found it impossible to grow single crystals of Zn+Cd by the C–G method when the Cd concentration was above 1.03 percent. Straumanis,[20] however, obtained specimens containing as much as 3 percent Cd which had all the appearance of single crystals and which he therefore called single. These were grown by the earlier method of Bridgman[21] and at the very low speed of 1.3 cm/hr.

The 0.26 percent Cd+Zn was studied also at the speed of 0.55 cm/min. The results are shown in Fig. 2. While the field was not explored as thoroughly as for the higher speed, it is clear that a range certainly exists although not the same range as before. The two most noticeable differences are first a shift of about 5° of the left-hand branch of the range and a widening out of the center branch sufficiently to join it definitely to the strip for θ near 90°. Lowering the speed, therefore, increases tremendously the possibility of growth of single crystals of this

[17] This was necessary since specimens of Cd pure zinc were mosaic and unsuitable for use as nuclei. It was also hoped by this use of nuclei containing Cd to start single crystals even when using pure zinc and thus discover a range of growth for it. This device, though successful with another method of growth (Poppy, reference 3) failed for the C–G method.

[18] Cd is soluble in Zn to the extent of 1 percent at room temperature. Circular of Bureau of Standards, No. 395, p. 140.

[19] P. Rosbaud and E. Schmid, Zeits. f. Physik **32**, 197 (1925). They do not state what speed of growth was used.

[20] M. Straumanis, Zeits. f. anorg. allgem. Chemie **180**, 1 (1929).

[21] P. W. Bridgman, Proc. Am. Acad. **60**, 308 (1925).

combination of Zn and Cd. These results are different from those of Hoyem and Tyndall who found that increasing the speed definitely raised the lower limit of their range. Nor are they in agreement with the hypothesis of Cinnamon[22] to the same effect, which, however, may not apply to impure zinc nor to growth of much smaller diameter crystals by another method at a different speed. The changes of orientation are similar to those for the higher speed, except that they are less in magnitude, rarely exceeding 7° or 8°.

Rosbaud and Schmid[19] and Straumanis,[20] in growing crystals of zinc with Cd impurity, found that the Cd entered the growing crystal in the same proportion in which it was present in the melt. This was found to be the case in the present work also.[23] They found, however, that the Cd did not distribute itself uniformly in the crystals but was segregated in layers parallel to the basal plane of the hexagonal lattice. This was evidenced in Straumanis's experiments by the appearance of uniformly spaced ridges on the surfaces of his crystals parallel to that plane. These appeared for Zn containing Cd (from mere traces up to 5 percent Cd[24]) but not redistilled and therefore presumably Cd free Zn. Etching experiments led Straumanis to the conclusion that the ridges were indeed richer in Cd than the layers between them.

The present writer finds the same periodic ridges[25] for Cd concentrations between 0.20 percent and 0.99 percent. They were prominent enough to be easily visible to the naked eye, and their prominence increased with the Cd content. Because of this and because they were always exactly parallel to the primary cleavage planes they made possible the determination of the orientation of the crystals before cleavage. By means of them it was quite easy also to follow the course of discontinuous and slowly continuous changes of orientation merely by examination of the crystal surfaces.

It seems quite certain in the light of the foregoing that the existence of the H–T range must be attributed mainly to the presence of the Cd. That the present work has not exactly duplicated it is probably due to the fact that other impurities besides Cd were present in the H–T zinc and undoubtedly exerted a modifying influence. It would be interesting to test this point directly by adding other elements besides the Cd in known amounts. This has been tried in a preliminary way only for Pb. The addition of only 0.02 percent Pb to 0.26 percent Cd+Zn apparently leaves the range unchanged, while 0.2 percent Pb very materially modifies it.

There is nothing in the above to disprove the hypothesis that crystals of pure (99.99 percent) zinc must have certain favorable growth conditions which are a function of the orientation. These conditions simply cannot be investigated by the C–G method[26] on account of the appearance of mosaics.

This work was done at the suggestion and under the direction of Professor E. P. T. Tyndall. For his very great help, constant interest and encouragement I thank him most heartily.

Most of the work was done in the writer's laboratory at Union College, Lincoln, Nebraska. Apparatus was loaned by the departments of Physics of the State University of Iowa and the University of Nebraska. I am also very grateful for this help.

[22] Cinnamon's hypothesis[3] states that the "ratio of the temperature gradient to the rate of growth must be near a certain favorable, but not necessarily the same, value for each orientation." According to this one should expect to find for lower speeds a range of successful growth at a lower temperature gradient, and therefore at a lower temperature in the present work in which $t-t_0$ is proportional to the gradient.

[23] By spectroscopic analysis carried out by Professor Tyndall.

[24] Straumanis's specimen containing 5 percent Cd was a polycrystal. However, each component showed the same typical periodicities in Cd segregation as did his "single" crystals containing 3 percent Cd or less.

[25] The fine drawings and photographs by Straumanis, some of which were reproduced recently in a paper by F. Zwicky, Rev. Mod. Phys. **6**, 193 (1934), illustrate completely and in detail the appearance of the present writer's crystals.

[26] That is, with any technique which the writer has tried so far. Given a way to eliminate mosaics there is no reason why the C–G method should not be used as a means of studying growth of single crystals of pure zinc.

Mathematical Physics of Metabolizing Systems with Reference to Living Cells

N. RASHEVSKY, *Department of Physiology, University of Chicago*

(Received January 7, 1935)

In previous papers we have investigated the conditions for spontaneous division of systems, which are seats of physico-chemical reactions *of any kind*. In this paper the previous general results are specialized for one particular set of reactions: oxidation of glucose and glycolysis. It is found, that whenever the two occur simultaneously, the ratio of their relative rates determines whether the system will divide spontaneously or not. Increased glycolysis favors spontaneous division. It is pointed out that the rapidly dividing cancer cells are characterized by an abnormally high rate of glycolysis.

IN a paper, published several years ago in this journal[1] we have shown how various general phenomena in living cells are in a natural way brought within the scope of studies of mathematical physicists. Such studies open a large field for physico-mathematical investigations, confronting us with problems, which being essentially physical in their nature, have nevertheless been very little, if at all, considered hitherto by physicists.

In subsequent papers[2–5] we have investigated mathematically various aspects of spontaneous division of small systems, which are seats of physico-chemical reactions. In the future we shall for brevity call such systems "metabolizing systems." Typical representatives of such systems are living cells. In fact metabolism is the *only physical* characteristic common to *all* growing cells,[6, 7] though they are of course more complex than the systems which we hitherto studied.

In calculating various factors which contribute to spontaneous division, we considered only such cases in which only one substance is being produced or absorbed in the system. When the system metabolizes a great number of different substances, the resulting effects are given by the sum of the contributions of all individual substances. The condition that *above a critical size* the system should become unstable and eventually divide is now expressed by

$$\sum_k q_k[\alpha(RT/M-\Gamma')/D_e-(\Gamma-\Gamma')/D_i]_k>0, \quad (1)$$

the sum being taken over all the metabolized substances. Eq. (1) replaces the simpler Eq. (47) of the former paper,[3] the notations being the same.

In the present paper we shall investigate a specific case of a set of reactions, frequently found in nature.

If we have a system in which all the reactions can be separated in two groups, one characterized by relatively large q_k's and the other by much smaller ones, we may *in the first approximation* extend the summation only over the first group. Such a situation actually is found in most living cells. For reactions connected with respiration $q\sim10^{-4}$ g·cm^{-3}·min.$^{-1}$. Those reactions include: Consumption of oxygen (O_2) and glucose (g); production of carbon dioxide (CO_2), lactic acid (l) and water (w). Considering the next important reactions, those connected with protein metabolism, we find $q\sim10^{-7}$ g·cm^{-3}·min.$^{-1}$. Others are still slower.

It is of special interest therefore to calculate the resulting effect of the five above-mentioned reactions. We do this by extending in (1) the summation over indices: 1, 2, 3, 4, 5, with the following meaning:

$$q_1=q_l>0; \quad q_2=q_{CO_2}>0; \quad q_3=q_w>0;$$
$$q_4=q_g<0; \quad q_5=q_{O_2}<0. \quad (2)$$

Indices of the other constants of (1) have correspondingly similar meanings.

Multiplication of *all* D_e and D_i with the same factor does not change (1); the same holds about

[1] Rashevsky, Physics **1**, 143 (1931).
[2] Rashevsky, Physics **2**, 303 (1932).
[3] Rashevsky, Physics **5**, 374 (1934).
[4] Rashevsky, Physics **6**, 33 (1935).
[5] Rashevsky, Physics **6**, 35 (1935).
[6] Philosophy of Science **1**, 176 (1934).
[7] Cold Spring Harbor Symposia on Quantitative Biology **2**, 188 (1934).

the q's. Hence we need to know only the relative values of the rates of reactions as well as of the diffusion coefficients.

Since the diffusion coefficient of the dissolved substance is determined by the resistance offered by the viscosity of the solute, and since approximately the latter may be calculated by the application of Stokes' formula,[8] considering the dissolved molecule as spherical, D must vary approximately as the inverse of the third root of molecular volume, which latter in its turn varies approximately as the molecular weight. We could of course introduce exact atomic volumes, but it would only somewhat complicate the calculations without changing the conclusions. The use of Stokes' formula is only roughly approximate, and does not justify greater exactitude in this next step. Substituting $D \sim 1/M^{\frac{1}{3}}$ into (1), and expressing, for reasons which will become obvious immediately, the relative rates of reactions not in $g \cdot cm^{-3} \cdot min.^{-1}$, but in *molecules** per cm^3 per min., we find

$$\sum_1^5 q_k[\alpha a(1-\Gamma' M/RT)M^{1/3} - b(\Gamma-\Gamma')M^{4/3}/RT]_k > 0, \quad (3)$$

a and b being coefficients of proportionality in the formulae, connecting D and $1/M^{\frac{1}{3}}$.

The five reactions considered here are connected together in the following way. Some molecules of glucose are simply split (glycolyzed) into lactic acid, according to the equation:

$$C_6H_{12}O_6 = 2C_3H_6O_3. \quad (4)$$

Some are oxidized to CO_2 and water, according to

$$C_6H_{12}O_6 + 6O_2 = 6CO_2 + 6H_2O. \quad (5)$$

It is customary to characterize the metabolism of a cell by the ratio β of the two reactions.[9] Then 3β represents the ratio of the number of glycolyzed molecules of sugar to the number completely oxidized.[9] If one molecule of glucose is oxidized, then 3β are glycolyzed. Altogether $3\beta+1$ glucose molecules are consumed. For the oxidation of one molecule 6 molecules of O_2 are necessary. 6β molecules of lactic acid and 6 molecules of CO_2 are produced.

Hence the relative values of the q's are:

$$q_1 = 6\beta;\ q_2 = 6;\ q_3 = 6;\ q_4 = -(3\beta+1);\ q_5 = -6. \quad (6)$$

An approximate estimation has shown us[3] that Γ and Γ' may be of the same order of magnitude as RT/M. A more exact calculation which will be given elsewhere, shows that

$$\Gamma = L/2M, \quad (7)$$

where L denotes the work necessary to bring one gram mole of dissolved substance from an ideal gas state into solution. A similar expression holds for Γ'.

Substituting (7) into (3) we find:

$$\sum_1^5 q_k[\alpha a(1-L'/2RT)M^{1/3} - b(L-L')M^{1/3}/2RT]_k > 0, \quad (8)$$

or, denoting the expression in square brackets by A_i and substituting (6) into (8):

$$(6A_1\beta + 6A_2 + 6A_3)/(3A_4\beta + A_4 + 6A_5) > 1, \quad (9)$$

which expresses the condition of spontaneous division of the system in terms of β.

For solutions of oxygen and CO_2 in water the L's can be calculated from the Bunsen partition coefficients and are found to be:

$$L_{CO_2} = -1{\cdot}7 \times 10^{10} \text{ ergs}; \qquad L_{O_2} = -3{\cdot}8 \times 10^{10} \text{ ergs}, \quad (10)$$

the minus sign indicating a force of repulsion between the molecules of the solvent and solute.[3] For the determination of L' no data are available at all. Nor do we know anything about L for glucose and lactic acid. Of the α's we know only that they are of the order of unity.

Putting $L_k = L_k'$, $\alpha_k = 1$, which should hold at least as a first approximation, and introducing (6) and (10), together with the numerical values of M's and RT into (8) we find:

$$(27\beta+45)/(17\beta+40) > 1. \quad (11)$$

The left-hand sides of (9) and (11) represent the ratio of the sum of the positive terms of (8),

[8] E. Baars, Geiger-Scheel Handb. d. Physik **13**, 437 (Springer, Berlin, 1928).

* That is substituting for q, qM/N, N—being Avogadro's number.

[9] O. Warburg, *The Metabolism of Tumors*, Constable and Company, London, 1930.

respectively, (1), to the sum of the negative ones. But this is nothing else[3] than the ratio of the sum of "dividing" forces, which satisfy Eq. (47) of the previous paper,[3] to the sum of the forces opposing division. From (11) we see that this ratio is the larger, the larger β. It is interesting to note in this connection that O. Warburg[9] has found that the abnormally easily dividing cancer cells are characterized by an abnormally high β. Whereas for cells of normal tissues β is practically zero, for cancer cells it is as high as 4. With $\beta=0$ the ratio (11) is 1·12; with $\beta=4$ it is 1·42.

It may be questioned whether water must be included amongst the substances produced in the system, if the latter itself consists mainly of water, as in the case of living cells. If the latter grow at such a rate as to double in about 10–24 hours, then all the water produced remains in the system to build it up and does not diffuse outwards. Such substances do not contribute to the osmotic and other forces.[1, 2, 3] Omitting the term corresponding to water in (8) and (9), that is putting $q_3=0$, we find

$$(27\beta+30)/(17\beta+40)>1, \qquad (12)$$

which is satisfied only if $\beta>1$. For $\beta=0$ no division can occur. For $\beta=4$, however, the dividing forces exceed the opposite ones by 28 percent.

If we consider only the effect of osmotic forces, that is if we put $\Gamma_k=\Gamma_k'=0$, then (8) leads to

$$(27\beta+21)/(17\beta+25)>1, \qquad (13)$$

which shows that for $\beta=0$ no division can occur at all. (13) is satisfied only for $\beta>0\cdot36$. But for $\beta=4$ the ratio (13) equals 1·39.

The physical reason for this can easily be visualized by remembering[3] that produced substances exert an osmotic pressure directed outwards and thus tend to divide the system into smaller parts. Consumed substances produce osmotic forces in opposite directions. Strong glycolysis results in an increase of *production* of lactic acid and in a relative decrease of consumption of oxygen.

The situation will not be changed materially by taking into account Γ for glucose and lactic acid. Though no direct data to calculate it are available, an indirect estimation of the effect of such a correction can be made. Glucose in solutions slightly *increases* the surface tension of water. Hence there is a force of attraction between molecules of glucose and of water. Γ is positive and its effect is therefore opposed to that of osmotic forces. It will tend, therefore, to decrease the "restoring" forces due to glucose. For lactic acid no data on surface tension are available, but from analogy to other similar compounds and from general theoretical considerations we should expect it to be *capillary-active*. This means a negative Γ and enhances the *dividing* forces produced by lactic acid. In any case, however, relations like (12), (11) or (13) are obtained.

We thus see that making *any* assumptions within a plausible range about various uncertainly known physical factors leads us to the same result, namely: Increased glycolysis leads to an increase of dividing forces and thus favors spontaneous division.

When (8) or (9) are satisfied, the critical size at which a spontaneous division occurs can be calculated in a similar manner as before[3] by equating the left-hand side of (1) to $8\gamma/R_0^2$. The values thus obtained are of the order of magnitude of 10^{-3} cm.

The calculations in this paper were based on (1) which holds for single-phase systems. However a similar expression is derived also from the formulae which we developed for two-phase systems,[4] and the general conclusions remain the same.

We have also studied the electric charges which are produced by metabolism.[5] Their possible effects on spontaneous division will be discussed elsewhere. Except for some special cases they are found to be of secondary importance as compared with those studied here.

Notice to Contributors

Manuscripts

Send manuscripts to John T. Tate, University of Minnesota, Minneapolis, Minnesota.

Papers must be in English, typewritten double spaced with wide margins.

Abstracts

An abstract must accompany each article. It should be adequate as an *index* and as a *summary*. As an index it should give all subjects, major and minor, concerning which new information is presented. As a summary it should give the conclusions of the article and all numerical results of general interest.

References

References should appear as footnotes, numbered consecutively, and arranged thus:

A. B. Smith, Phys. Rev. **41**, 852 (1932).

Captions and Legends

Legends for figures must accompany manuscript. Each table should have a caption. These in all cases should be complete in themselves so as to make the data intelligible to the reader without reference to the text.

Mathematical Expressions

Make all expressions clear to the typesetter. Identify in the margin of the manuscript Greek letters and unusual symbols.

Use fractional exponents instead of root signs.

Avoid complicated exponents and subscripts. Should it be necessary to repeat a complicated expression a number of times it should be represented by some convenient symbol.

The solidus (/) should be used wherever possible for fractions.

Illustrations

Plan figures for approximately one-half reduction. Wherever possible arrange figures so that after reduction they will not be wider than three inches. Lettering must be large enough to be readable after reduction.

Line drawings must be made with India ink on white paper or tracing cloth. Coordinate paper is not desirable, but if used must be blue-lined and all coordinates to be reproduced drawn with India ink.

Drawings which cannot be used will be returned to the author or redrawn at his expense.

Photographs should show clear black and white contrasts.

Avoid round and oval photographs.

Publication Charge

A portion of the cost of publishing the results of research must be regarded as an integral part of the cost of the research. In keeping with the policy which is effective for the Institute's publications a bill for $3.00 per page will be sent to the authors of papers appearing in this journal. It is intended that the authors shall present these bills for payment to the institutions which have supported their researches. If the institution is unable to make payment the bills are to be returned marked "No Funds Available."

Reprints

As a partial return for the publication charge one hundred reprints without covers will be furnished free. Additional reprints may be obtained at a regular schedule of prices.

Alterations

Take great care that each manuscript is typographically accurate. A limited number of alterations in proof are unavoidable, but the cost of making extensive alterations after the article has been set in type will be charged to the author.

Proof and all correspondence concerning papers in the process of publication should be addressed to the Editorial Secretary, American Institute of Physics, 11 East 38th Street, New York, N. Y.

THE AMERICAN PHYSICAL SOCIETY

R. W. WOOD, *President*
The Johns Hopkins University
Baltimore, Maryland

F. K. RICHTMYER, *Vice President*
Cornell University
Ithaca, New York

W. L. SEVERINGHAUS, *Secretary*
Columbia University
New York, New York

G. B. PEGRAM, *Treasurer*
Columbia University
New York, New York

JOHN T. TATE, *Editor*
University of Minnesota
Minneapolis, Minnesota

L. B. LOEB, *Local Secretary for the Pacific Coast*
University of California
Berkeley, California

THE object of the American Physical Society is the advancement and diffusion of the knowledge of physics. It sponsors *The Physical Review*, *Physics* and the *Reviews of Modern Physics*. Upon the recommendation of two or more members, any person engaged in work in the field of physics or actively interested in physics may become a member; receive one of the journals; receive also *Science Abstracts* A, *The Review of Scientific Instruments with Physics News and Views*, and the Bulletin of all meetings; and attend and participate in the seven annual meetings.

Annual dues: $10. Fellows, $14.

For further information address the Secretary as above.

THE SOCIETY OF RHEOLOGY

S. E. SHEPPARD, *President*
Eastman Kodak Company
Rochester, New York

EUGENE C. BINGHAM, *Editor*
Lafayette College
Easton, Pennsylvania

A. STUART HUNTER, *Secretary & Treasurer*
Du Pont Rayon Company
Buffalo, New York

THE Society of Rheology was formed in 1929 to advance the science of the deformation and flow of matter. It cooperates with the American Physical Society in editing *Physics* of which four issues per year are devoted to *Rheology*. All persons interested are invited to become members and participate in the proceedings at the annual December meetings. Members receive without other charge than their dues all issues of *Physics*, *The Review of Scientific Instruments with Physics News and Views* and the notices of the Society.

Annual dues: $6.

For further information address the Secretary as above.